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NATURE OF THE RADIOACTIVE DEBRIS AND NUCLEAR RADIATION ASSOCIATED
WITH AN UNDERWATER NUCLEAR EXPLOSION (U)

USNRDL-TR

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Folder Nature of the Radioactive Debris

L. B. Werner

.... Nuclear Explosion USNRDL-TR

27 January 1964

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ABSTRACT

A technical review on the nature of the radioactive debris and nuclear radiation from underwater nuclear explosions is presented for future inclusion as Chapter 10 of the planned DASA book Underwater Nuclear Explosions, Part 1 - Phenomena.

The nuclear processes considered include fission, fusion, radioactive decay and neutron capture. Both those processes which determine the nature of the explosion itself and those which result in the production of nuclear radiation fields are discussed. Factors, which influence the radiochemical composition of the debris and the nature and measurement of ionizing radiation are presented.

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SUMMARY

The Problem

To condense into one chapter, the basic concepts of the nature of the radioactive debris and associated nuclear radiations from an underwater nuclear explosion. This chapter to be part of the DASA book on Underwater Explosions, Part 1 - Phenomena.

Findings

A thorough search of the literature was made and from these documents a brief technical description of the debris and radiation was summarized. This description was constructed not as a comprehensive review but as a brief, from which further studies can be made.

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FOREWORD

This report was prepared for inclusion as Chapter 10 of the planned DASA book on Underwater Nuclear Explosions, Part 1 - Phenomena. The completed volume will consist of the following:

- Chapter 1 - Introduction
- 2 - Hydrodynamic Considerations
- 3 - Theory of Similitude
- 4 - The Shock Wave
- 5 - Shock Wave *interactions* Propagation Under Special Oceanographic Conditions
- 6 - The Explosion Bubble
- 7 - Underwater Cratering
- 8 - Surface Waves
- 9 - Surface Phenomena
- 10 - Nature of the Radioactive Debris and Nuclear Radiation.
- 11 - Distribution of the Radioactive Debris and Nuclear Radiation

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UNDERWATER NUCLEAR EXPLOSIONS

PART I - PHENOMENA*

CHAPTER 10

NATURE OF THE RADIOACTIVE DEBRIS AND NUCLEAR RADIATION

An appreciation of the important aspects of underwater nuclear explosions requires an understanding of some physical-chemical and nuclear phenomena which will be discussed in this chapter. The nuclear processes to be considered include fission, fusion, radioactive decay, and neutron capture. Two aspects of the operation of these processes may be delineated: those which determine the nature of the detonation itself and those which result in the production of a nuclear radiation hazard. (S)

The energy released per unit volume in a nuclear explosion is very large, but there are not correspondingly large quantities of explosion products produced. This engenders some unique features of nuclear detonations which are discussed elsewhere in this book. Attention is given in this chapter to the formation of radioactive products and to radiation whose effects may extend beyond the range of physical damage from the explosion. Some factors which influence the radiochemical composition of debris and the nature and measurement of ionizing radiation are also presented. (S)

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* See Foreword.

~~** Security classification is indicated for individual paragraphs.~~

10.1 FISSION, FUSION, AND THE RADIOACTIVE PRODUCTS OF BOMB DEBRIS

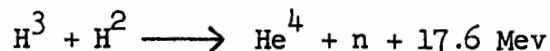
The detonation of all nuclear weapons depends basically upon the occurrence of a fission chain reaction in which the nuclei of certain isotopes of elements of high atomic number split into two smaller fragments called fission products. The process is accompanied by the average emission of two or more neutrons and the liberation of a quantity of energy which is many orders of magnitude larger on an equal mass basis than that for ordinary chemical reactions such as the detonation of TNT. The fission process is energetically possible for a large number of heavy nuclides, but for most of these it does not take place at an appreciable rate unless disturbing energy is provided the nucleus. In fission chain reactions this "energy" is derived from the absorption of a neutron, although other means of experimentally inducing fission exist. (S)

It has been observed that the sum of the rest masses of all the products of fission is less than the sum of the rest masses of the fissioning nucleus and absorbed neutron. This difference in rest mass appears as energy released in fission, in accordance with Einstein's equivalence of mass and energy ($E=mc^2$). The mass-energy equivalence is found in the concept of "mass defect." The rest mass of a nucleus is found to be less than the sum of the masses of the constituent neutrons and protons. This difference, called the mass defect, is equated to the nuclear binding energy of the nucleus. This is the energy which would be released by neutrons and protons combining to form a given nucleus, or, conversely, which would be necessary to separate the nucleus into individual neutrons and protons. The mass defect, i.e., the binding energy, for elements near the middle of the periodic table is greater than that for light elements or for heavy elements, including the fissionable isotopes. For this reason the splitting, i.e., fission, of heavy elements into lighter fragments is energetically favorable. Also for this reason another type of reaction is energetically favorable, viz., thermonuclear reactions (fusion), which involve the combination of light elements into heavier ones. (S)

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The critical requirement for the initiation of thermonuclear reactions is the achievement of very high temperatures of the order of millions of degrees. This provides the nuclei with sufficient thermal energy to overcome electrostatic repulsive forces and approach closely enough (of the order of 10^{-13} cm) for combination to occur. These high temperatures can be achieved by detonation of a fission device. Once begun the thermonuclear reaction

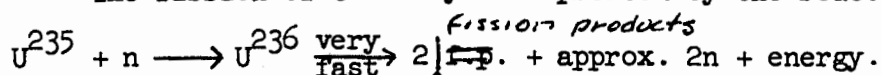
can be self-propagating and result in explosions of very high yield. One important thermonuclear reaction is the fusion of two hydrogen isotopes to form helium by the reaction



The neutrons liberated in such reactions possess considerable kinetic energy (14 Mev) and may be utilized efficiently to induce further fission in U^{235} , Pu^{239} or in U^{238} , the latter of which does not fission with low energy neutrons. Fission devices which incorporate fissionable material for this purpose are referred to as "boosted weapons." In this case the thermonuclear reaction provides some additional energy to the detonation but the main effect in boosted weapons is to increase the yield due to fission. For weapons that contain a U^{238} tamper, that is, a reflector to reduce escape of high energy neutrons from the core of the weapon, fission may be induced in the tamper by the high energy neutrons from the thermonuclear reaction. ~~✗~~

10.1.1 THE FISSION PROCESS

The fission of U^{235} may be expressed by the reaction



The number of neutrons emitted varies, but the average number has been found to be 2.5 for U^{235} and 3.0 for Pu^{239} . ~~(✗)~~

Once fission is begun, the neutrons emitted are reabsorbed in about 10^{-8} sec for successive generations of fissions. If the amount of fissionable material is above a certain minimum or "critical" mass, the generation of neutrons exceeds the loss of neutrons through escape and through captures not resulting in fission, and a chain reaction is established. Eventually the rate of neutron loss exceeds the rate of neutron production, and the reaction stops, before all of the fissionable nuclei have fissioned. The ratio of number of fissions to number of fissionable nuclei is a measure of the "efficiency" of the weapon. ~~(✗)~~

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The mechanism of fission was described by Bohr and Wheeler on the basis of the liquid-drop model of the nucleus. The nucleus may be visualized as being held in a normally spherical configuration by nuclear forces between the constituent nucleons (protons and neutrons), analogous to surface tension holding a drop of water in a spherical shape. When energy is added to the nucleus, i.e.,

by absorption of a neutron, deformations can occur, one form of which resembles a dumbbell. In this state the electrostatic repulsion by the positively charged ends may prevent return to a spherical shape and fission occurs. The process is illustrated in Fig. 10.1:1 as stages A through D.

The theory relates the ease of fission with the value of Z^2/A for the nucleus where

Z = nuclear charge (atomic number or number of protons in the nucleus)

A = mass number (number of protons plus neutrons in the nucleus)

Less activation energy is required for high values of Z^2/A . Once formed, the fission products move apart with a high kinetic energy due to electrostatic repulsion. The fission neutrons emitted also possess kinetic energy, so that the instantaneous release of energy (rated yield) is about 177 Mev per fission. Further energy release occurs in the radioactive decay of fission products so that the total energy release is about 200 Mev as shown in Table 10.1:1 as taken from Glasstone (1962).

Table 10.1:1

Distribution of Fission Energy

	Mev
Kinetic energy of fission fragments	165 \pm 5
Instantaneous gamma-ray energy	7 \pm 1
Kinetic energy of fission neutrons (average value)	5 \pm 0.5
Beta particles from fission products	7 \pm 1
Gamma rays from fission products	6 \pm 1
Neutrinos from fission products	10
Total energy per fission	200 \pm 6

Since one atomic mass unit is equivalent to about 931 Mev or 1.66×10^{-24} gm, the total energy per fission is equivalent to 0.357×10^{-24} gm. For a nuclear yield equivalent to 1 kt of TNT (by definition 10^{12} calories), it may be calculated that complete fission of about 53 gms of fissionable material is required. *(10)*

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One of the most interesting features of the low energy fission process is the lack of symmetry of its products, i.e., the two nuclear

Fig. 10.1:1 Mechanism of Fission According to the Liquid-Drop Model of the Nucleus

fragments have unequal masses. Fission results in a broad distribution of mass numbers among the fission products, and, similarly, a large number of chemical elements is produced. (X)

10.1.2 PRODUCTS FORMED IN FISSION

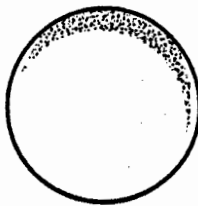
The relative number of fissions that result in a particular fission product is called the "fission yield" of that product, expressed either as a percentage of the total, or as a number of atoms of a fission product for a given number of fissions. A semilogarithmic plot of fission yield vs. mass number is shown in Fig. 10.1:2. (X)

Examination of the curve shows that the products fall into two broad mass-number groups, a "light" group with mass numbers from about 85 to 104 and a "heavy" group with mass numbers from about 130 to 149. The most probable type of fission may be seen to consist of division into two fragments having masses about 95 and 139. Similar curves are obtained for all fissioning nuclei, although for Pu^{239} fission for example, the peaks are shifted to slightly higher mass numbers. For fission caused by higher than thermal ^{neutron} energies, such as bomb neutrons, the peaks are spread slightly and the valley, which represents symmetrical fission, is raised. (X)

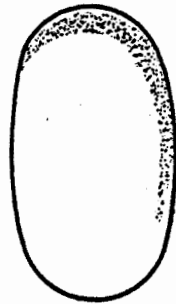
The way in which the protons in the compound nucleus are divided between the fission fragments is also important. For U^{236} , $Z/A = 92/236$. If this ratio were preserved in the fission fragments, the most probable type of fission would have 37 and 55 protons in the two fragments, corresponding to the elements rubidium and cesium respectively. Actually, the number of protons associated with a given mass number is not constant among the primary fission products so that several elements may

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DD FORM



A



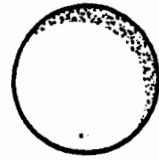
B



C



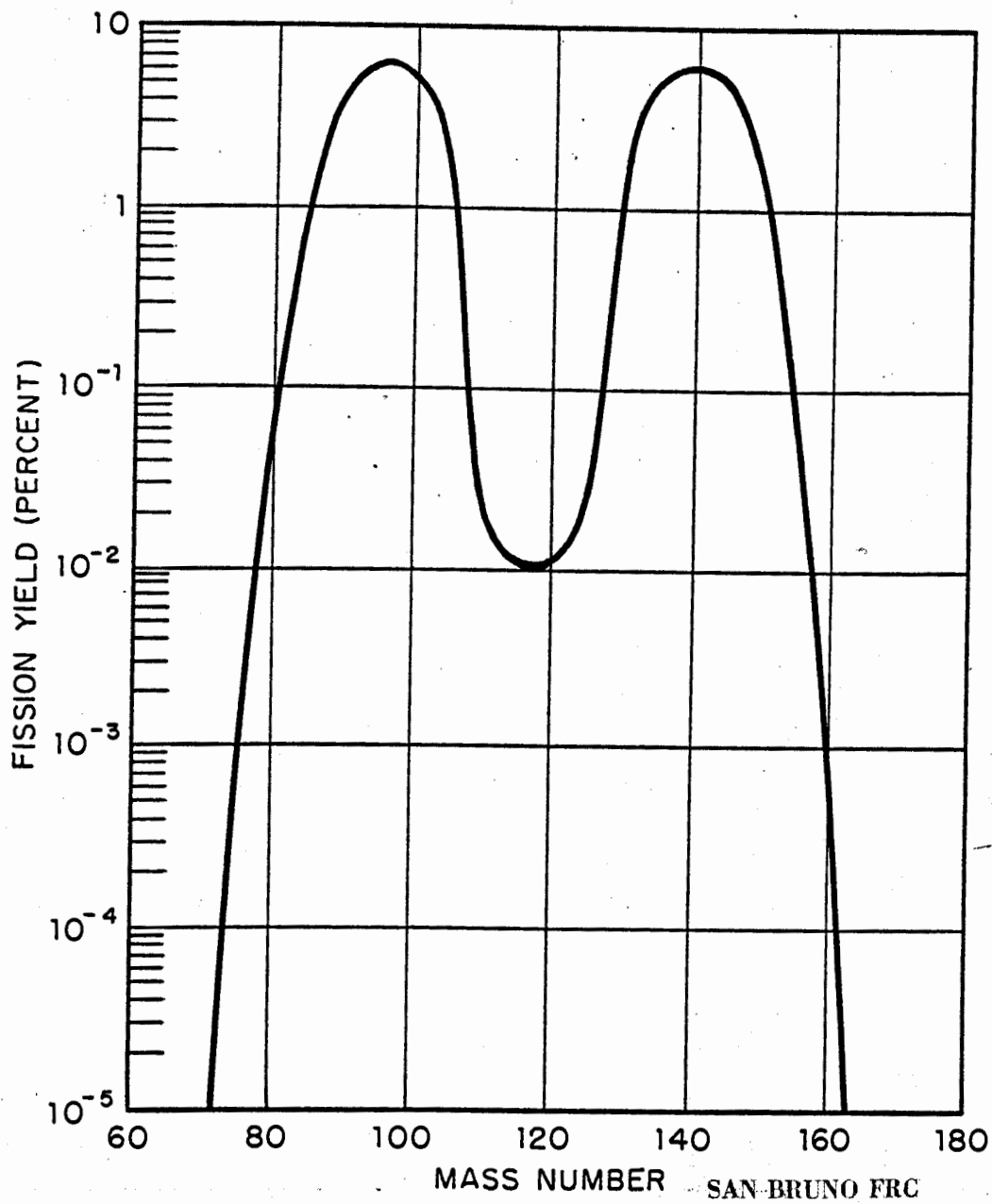
D



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Fig. 10.1:2 Fission-Yield Curve for U^{235} and Slow Neutrons

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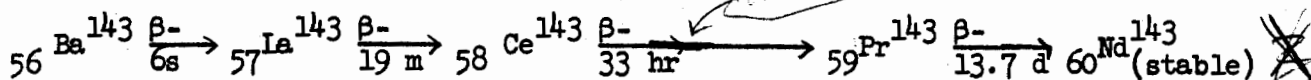
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be formed with identical mass numbers. The low ratio of protons to neutrons which the fission product inherits from the compound nucleus leads to nuclear instability. To reach stability the primary fission products emit neutrons (instantaneous and in some cases delayed) and then undergo radioactive decay by beta particle emission, with a resulting increase in Z. This process continues until a stable charge-to-mass ratio is reached. For example,



The net result is that over 36 different elements are produced directly in fission or by subsequent beta decay. The relative proportions of these elements change with time after fission, and the total number of different nuclides produced exceeds 200. Figure 10.1:3 shows a number of important decay chains. (For a comprehensive description of the U^{235} fission products, the reader is referred to Bolles and Ballou.)

10.1.3 CHEMICAL AND PHYSICAL STATE

The state of the fission products following their formation is determined by the nature of the explosion and the environment in which the detonation takes place. The post-shot transport and deposition of the fission products, as well as their contamination-decontamination behavior and availability to biological systems, are functions of their chemical and physical state.

Initially, all constituents of the fireball in an underwater detonation may be visualized to be vaporized and highly ionized. As this system cools to condensation temperatures, nucleation and particle formation occur. The initial chemical form of the particles is determined by the components of the fireball and the resulting steam bubble and by the thermodynamic and kinetic features of the reactions which can occur. Eventually very substantial quantities of environmental materials, e.g., seawater, become mixed with the primary bomb debris and react with it.

Attempts were made to predict on theoretical grounds both the chemical composition and particle size for tower, land-surface, and barge shots on water (see Magee, Adams et al. Mackin et al., and Miller (1950)) although relatively little work was done for the underwater case.* However, Bunney, et al., made estimates of the

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*These processes are discussed further in 10.3.3, in relation to fractionation phenomena.



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Fig. 10.1:3 Several Important Fission Product Decay Chains. (Half-lives in parentheses; asterisks indicate estimated values.)

Mass
No.

Decay Chains

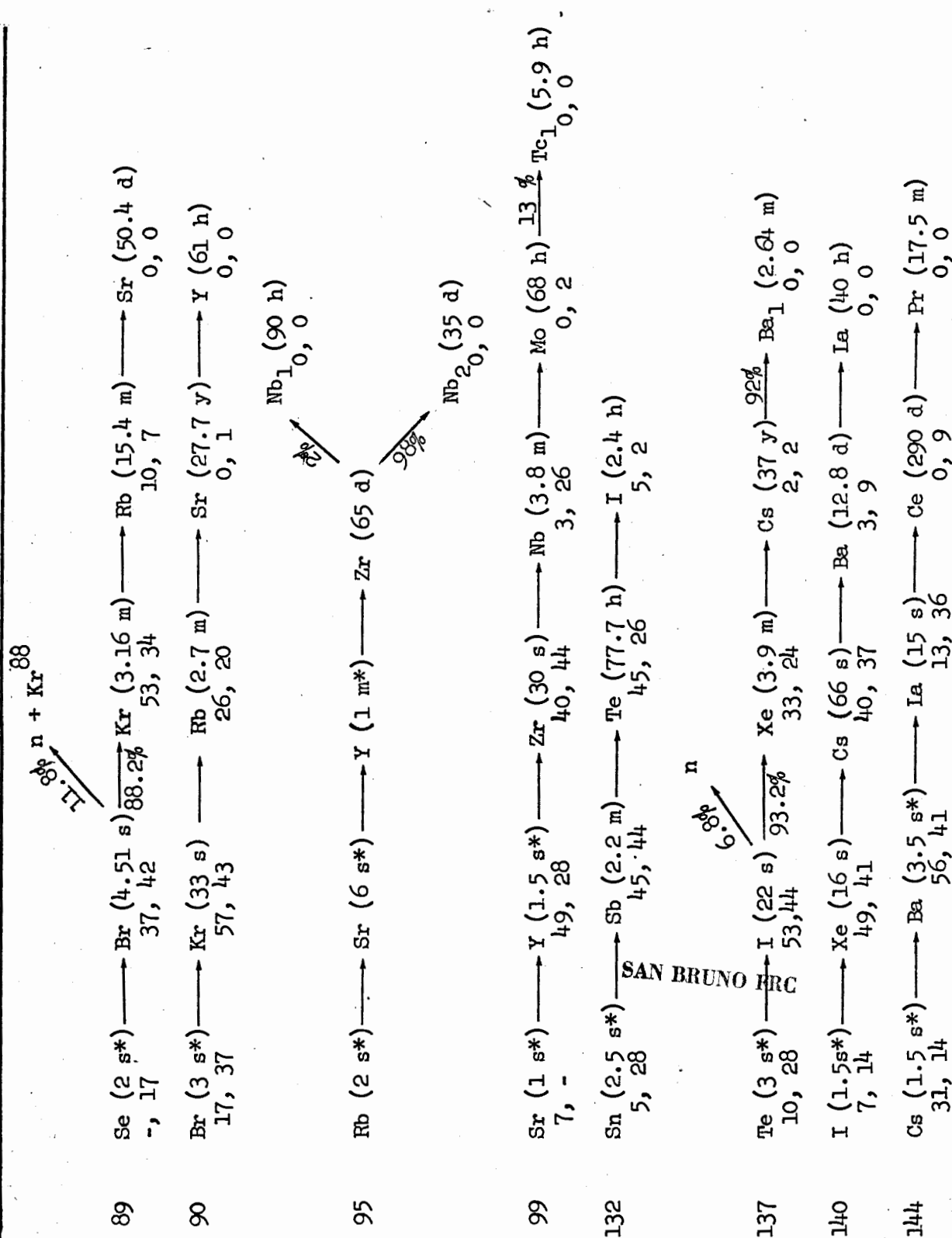


Fig 10.1.3

of the predominant chemical species from a shallow underwater burst, based upon available thermodynamic data. Oxides and chlorides are regarded as the most important intermediate species, but hydration is also considered to be important although it is more difficult to predict because the partial pressure of water vapor varies with the condition of the detonation and knowledge is lacking about rates of reaction. The final state of the fission products in liquid water will depend upon these factors and upon the solubilities of the compounds in seawater. The state may change with time owing to slow rates of dissolution, and to adsorption of dissolved materials on the surfaces of solids which are in contact with contaminated seawater. For convenience, the chemical elements may be placed into groups according to their chemical similarities, e.g., rare gases (Kr and Xe); alkali metals (K, Na, Cs, Rb), etc. An estimate of the most likely physical state for various groups of chemical elements derived from bomb debris and seawater is given in Table 10.1:2. (S).

Some measurements were made of the physical state of bomb debris from underwater detonations by Ballou and of the state of elements in laboratory underwater electrical arcing experiments by Freiling, et al. and Ballou (1962). Gross agreement between estimated and observed states was obtained, although certain elements ~~show a greater tendency toward solubility~~ than predicted, particularly in the absence of solids. Further studies of the chemical and physical state, as related to variations in conditions of detonation and the seawater medium, are definitely required. (S)

10.1.4 NATURE OF NUCLEAR RADIATION

In the previous section on fission and the fission products, reference was made to neutron emission and to beta and gamma rays which accompany radioactive decay. The properties of each type of radiation and its interaction with matter will be described in this section. In addition, alpha emission, which is the normal mode of radioactive decay for U²³⁵, U²³⁸ and Pu²³⁹ will be discussed briefly. (S) SAN BRUNO FRC

Alpha particles are identical with the positively charged nuclei of helium atoms. They are composed of two neutrons and two protons and weigh about 6.64×10^{-24} gms. Alpha emission occurs spontaneously only among elements heavier than lead (atomic number 83), except for several isotopes of the rare earth elements. The energy with which alpha particles are emitted is relatively high (about 4 to 9 Mev), but in spite of this they are easily stopped by relatively thin layers of absorbing material. The stopping process may be visualized as the interaction of the electric field surrounding the alpha particle with the electrons of atoms of the absorbing medium. Part of the kinetic energy of the alpha

Table 10.1:2

Estimated Predominant Physical State for Constituents of Bomb Debris and Seawater

Chemical Group	Principal Elements in Bomb Debris	Principal Elements in Seawater	Predominant Physical State
Rare gases	Kr, Xe	-	gas
Halogens	Br, I	Cl	ionic
Alkali metals	Rb, Cs	Na, K	ionic
Alkaline earths	Sr, Ba	Mg, Ca	ionic
Rare earths	Y, La-Tb	-	solid
Oxygenated anions	As, Se, Mo, Tc, Te	S	ionic (Te, solid)
Niobium-zirconium	Nb, Zr	-	solid
Noble Metals	Ru, Rh, Pd	-	solid
Miscellaneous	Ni-Ge, Ag-Sb, Fe	-	solid (Ge, ionic)
Heavy elements	U, Np	-	solid

particle is transferred to the electrons, sometimes knocking the electron out of the atoms to which it was bound and thereby producing ionization of the medium. For a particle whose velocity is much less than the speed of light, the rate at which energy is lost along its path is given approximately by

$$-\frac{dE}{dx} \propto \frac{Mz^2}{E^2} (NZ) \quad \text{Large } Z$$

$$-\frac{dE}{dx} \propto \frac{Mz^2}{E} (NZ) \quad \text{Small } Z \quad (10.1:1)$$

where z = charge of the moving particle

M = mass of the moving particle

E = energy of the moving particle

NZ = Number of electrons/cm² in the medium

Examination of this equation shows that the rate of energy loss actually increases as the particle loses energy. The maximum density of ionization, therefore, takes place near the end of the particle's range (Fig. 10.1:4).

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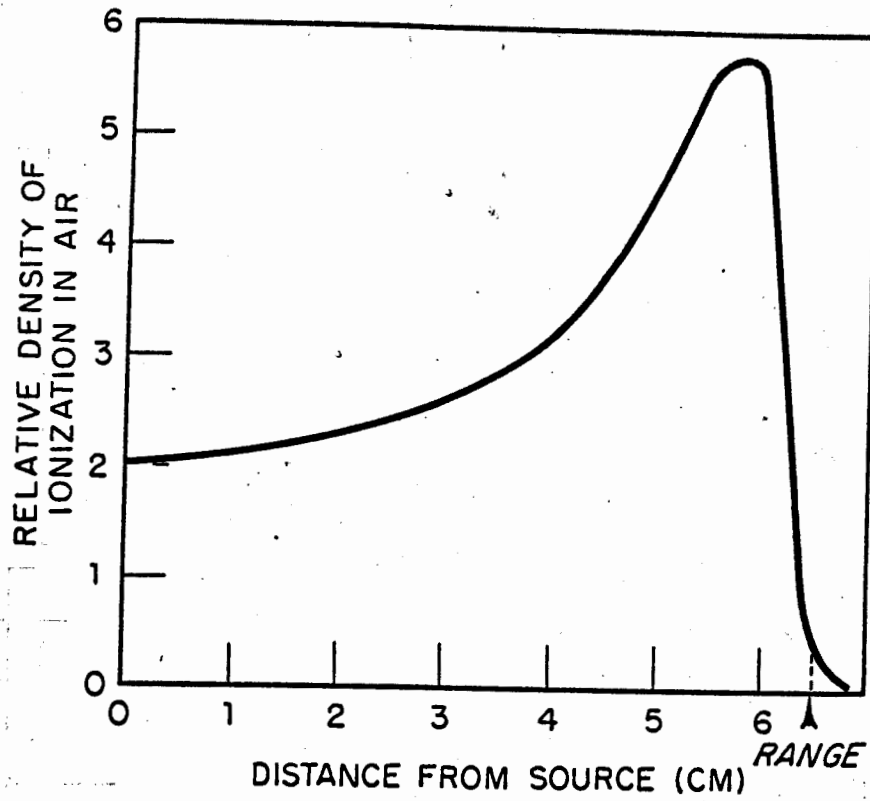
Because of the limited range of alpha particles in matter, thin material such as paper or epidermis normally is sufficient to absorb the radiation completely. This accounts for the negligible biological hazard from alpha radiation, unless the source is taken inside the body where there is intimate contact with radiation-sensitive tissues. It

Fig. 10.1:4 Relative Density of Ionization at Various Distances
From a Source of Alpha Particles

also accounts for the difficulty in measuring alpha radiation accurately. Detectors with very thin windows must be used in close proximity to the source, and account must be taken of self-absorption of alpha particles by the sample itself. In field work the presence of significant amounts of inert absorbing material is practically inevitable, and large errors can be introduced in the measurement of alpha source intensities. (S)

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Beta radiation also consists of the emission of charged particles by nuclei. The particles have unit negative charge and a rest mass equal to only about 9.1×10^{-28} gm. They are designated by the symbol β^- and are identical with ordinary free electrons. Radiation of positively charged beta particles or positrons also occurs among certain artificially produced radionuclides, but since none of the fission products or fissionable elements decay by this process, it will not be considered further in this chapter. Beta particles like alpha particles also transfer energy to electrons and produce ionization in an absorbing medium. But because of its much smaller mass, the beta particle may lose much larger fractions of its energy in a single encounter and is subject to large deflections in its path. The range of beta particles in air is greater than that of alpha particles, being of the order of



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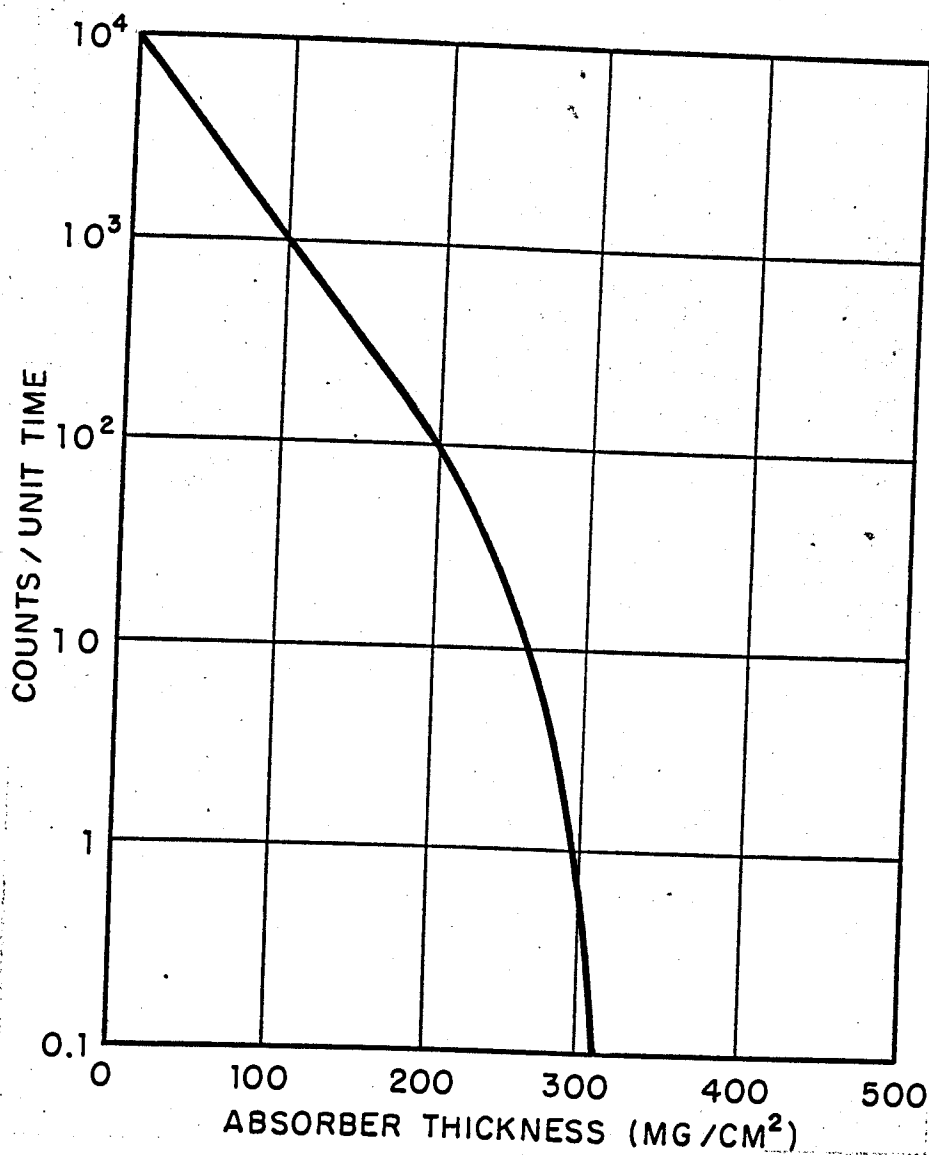
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Fig. 10.1:5 Typical Beta Absorption Curve

hundreds of centimeters. Beta particles are not emitted monoenergetically, but have a ^{continuous} distribution of energies between zero and a maximum value characteristic of the emitter. The absorption of betas tends to vary exponentially with the thickness of the absorbing material, except at the maximum end of their ^{path.} range (Fig. 10.1:5).

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Another mechanism for loss of beta energy consists of the radiation of electromagnetic energy as ^{the beta particle} it passes near the nucleus of an atom and is accelerated toward the nucleus by electrostatic attraction. The radiation which results from this acceleration is called bremsstrahlung and has the same properties as soft X rays. It is commonly observed but is an important means of energy loss only for high energy electrons.



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than do alpha or beta particles. Since neutrons have no electric charge there is no scattering in the medium due to Coulomb forces. This greatly increases their penetrating power.

Since moderate amounts of absorbing material will shield beta rays, they are only detected efficiently by instruments having unshielded detectors which are held close (i.e., less than the maximum range in air) to the source. However, in contrast with alpha rays, beta rays can cause severe radiation effects on epidermis and underlying shallow layers of tissue if the source is close to or in contact with the body.

Neutron radiation behaves very differently with respect to interactions with matter ~~since neutrons have no electric charge~~. Neutrons interact with matter in two principal ways: by collision and scattering of the neutron, and by neutron capture. Collisions between neutrons and nuclei of the medium through which they pass results in a progressive transfer of the neutrons' energy to the medium and eventual slowing of the neutrons to energies determined purely by the temperature of the medium, i.e., to thermal energies. The collision may be of the "billiard-ball" type where the sum of the kinetic energies of the neutron and the nucleus after collision equal the kinetic energy of the neutron before collision. This is called elastic scattering. Energy loss is greatest by this process when the collision takes place with a light nucleus, hydrogen being the most effective of all neutron energy moderators. If, however, part of the neutron's kinetic energy leads to excitation of the target nucleus the process is referred to as inelastic scattering. Thermal neutrons continue to diffuse through the medium until captured. The stopping power of a neutron absorber thus is a function of its effectiveness as a moderator and its cross-section for neutron capture.

Neutrons, of course, do not produce ionization directly but may do so through recoil of target nuclei, especially hydrogen. Ionization is also produced by gamma rays which accompany neutron-capture reactions. These effects, combined with the penetration power of neutrons, make neutron radiation a distinct biological hazard. Its importance relative to initial gamma radiation from a detonation, depends on yield, distance, and shot conditions. For small yields at close distances from above-surface shots neutron radiation predominates over initial gamma radiation.

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Of the nuclear radiations being considered, gamma radiation is distinctly different both in nature and in its interaction with matter. In its properties, gamma radiation is identical with X rays since both are electromagnetic radiation. Gamma rays, however, originate in the nucleus as a result of electric or magnetic multiple oscillation, namely, by movements of electric charges within the nucleus. X rays originate in the electronic structure of the atom. These processes are quite analogous to radiation of radio waves from antenna. The energy of gamma rays is usually greater than that of X rays. The wave length of gamma radiation is thus usually shorter than that of X rays, and gamma rays generally also are much more penetrating. Individual emission by a nucleus results in release of a quantum of electromagnetic energy called a photon.

The absorption and scattering of gamma rays occurs mainly by interaction with the atomic electrons according to

$$I = I_0 B(x) e^{-\mu x} \quad (10.1:2)$$

where I_0 = initial intensity of a beam of gamma rays
 I = intensity of gamma rays after passing through an absorber
 μ = linear attenuation coefficient (cm^{-1})
 x = distance (cm) through absorber
 $B(x)$ = build-up factor

The linear attenuation coefficient which accounts for the effect of photon absorption in the medium and scatter out of the beam is a function of both the medium and the gamma ray energy. When calculating the amount of energy actually deposited in a volume element of the absorber (Dose), the linear absorption coefficient, which may be substantially smaller than the total attenuation coefficient, is used in equation (10.1:2). A useful concept is the "half-thickness," i.e., the thickness of absorber which reduces the intensity to one half the initial value. From equation (10.1:2) the half-thickness is seen to be $\log_2 e / \mu = 0.693/\mu$. The absorber thickness may also be expressed in gm/cm^2 , in which case μ becomes the "mass attenuation coefficient" and the following relationship applies

$$\mu \text{ (mass attenuation)} = \frac{\mu \text{ (linear)}}{\rho} \frac{\text{cm}^2}{\text{gm}} \quad (10.1:3)$$

where ρ = density of the absorbing material. (N)

Three mechanisms exist which account for absorption of gamma rays in matter: the photoelectric effect, the Compton effect, and pair production. At low gamma energies the principal mechanism is the photoelectric effect in which an interaction with atomic electrons results in ejection of an electron from its orbit and disappearance of the photon. The kinetic energy of the electron is the difference between the photon energy and the energy with which the electron was bound in its orbit. Electron ejection cannot take place unless the energy of the gamma photon equals or exceeds the electron binding energy. For this reason, the absorption coefficient for a given element shows sharp discontinuities at this binding energy when plotted against gamma energy. Since each element differs in its electron binding energies, the discontinuities, i.e., peaks in the absorption coefficient curve, vary from element to element and appear at the highest energies for elements of high atomic number. Since more than one type of orbital electron may be involved in this process, more than one peak appears for each element. (N)

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~~At intermediate gamma energies (0.1 to 10 Mev) the Compton effect is the principal means of gamma energy loss. Only a portion of the photon's energy is transferred to an electron, and a scattered photon~~

At intermediate gamma energies (0.1 to 10 Mev) the Compton effect is the principal means of gamma energy loss. Only a portion of the photon's energy is transferred to an electron, and a scattered photon of reduced energy results. For broad radiation beams and thick absorbers, such as are of interest in shielding from nuclear explosions, the Compton photon may be scattered several times before emerging from the shield or absorbed by the photoelectric process. When this occurs photons originally lost can be scattered back into the gamma ray beam and I, the intensity of the emerging radiation, is larger than that given by pure attenuation alone. Allowance for this multiple scattering is made by including a "build-up factor", represented by $B(x)$ in equation (10.1:2). The value of $B(x)$ depends upon the thickness of the absorber, the nature of the material, and the energy of the gamma ray beam. Also, the direction of the photons change, which can produce substantial radiation intensities at locations which are shielded from line-of-sight contact with the source. (N)

Since momentum as well as energy is conserved, because it is highly penetrating, and capable of producing ionization through interactions with electrons, gamma radiation is most important in radiological effects of underwater detonations. Emission of gamma radiation accompanies nearly every type of nuclear reaction, including neutron capture, fission, and beta decay. (N)

10.1.5 ENERGY SPECTRA

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In discussing the various types of radiation, it has been noted that beta particles are emitted with a continuous energy spectrum. Fission neutrons also possess a continuous spectrum, although of a different type. In gamma emission, however, the photon energies are discrete and are characteristic of the particular emitter. In general a nucleus emits more than one wave length of gamma radiation; this is related to the multiplicity of nuclear energy levels which can exist within the nucleus. A complete nuclear description can be given only by reference to quantum mechanical theory, which is outside the scope of this chapter. But the essential nature of the situation may be visualized with the aid of an energy-level diagram. An example of such a diagram in which a number of excited nuclear states and the transitions which occur between them are represented in Fig. 10.1:6, as taken from Miller (1957a). In addition to the ground state, or zero energy level, six excited energy states are shown for Sr^{89} . Beta decay of Rb^{89} may leave the product nucleus Sr^{89} in the ground state or in an excited state from which the nucleus drops by one or more emissions of a gamma photon to the ground state. The greatest maximum beta energy, viz., 3.92 Mev, occurs with beta decay directly to the ground state. The most frequent beta decay is to the 2.31 Mev state for which the maximum beta decay energy is 1.61 Mev. The relative abundance of this transition is about 0.5, i.e., about half of all beta disintegrations go by this path. Certain transitions between energy states are not observed or have a very low relative abundance.

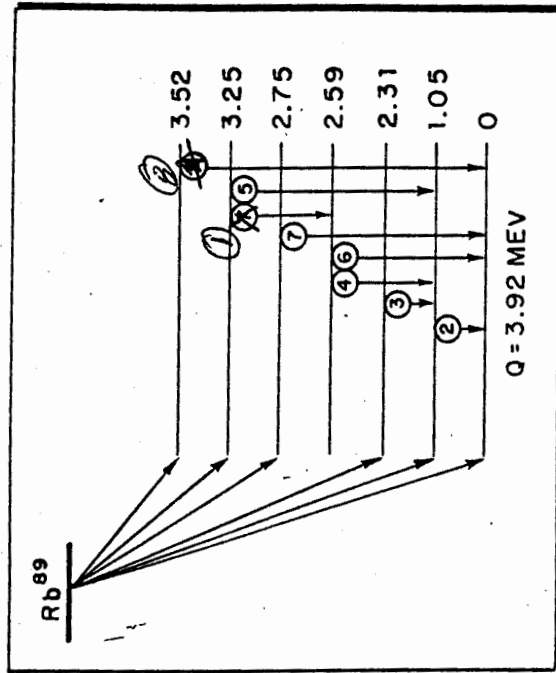
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Fig. 10.1:6 Energy Level Diagram and Decay Scheme for Rb^{89} - Sr^{89}

Nuclide: Rb^{89} (14.9m)

Max. Beta Energy Relative Abundance

0.40	0.021
0.67	0.289
1.17	0.026
1.61	0.508
2.87	0.066
3.92	0.090



Photon Abundance per 100 dis

No.	E_g	N_g	α	N	N_k	$N E_g$ (MeV/dis)
1	0.663	15.4	< 0.001	15.4	-	0.1021
2	1.05	71.0	< 0.001	71.0	-	0.7455
3	1.26	50.9	< 0.001	50.9	-	0.6415
4	1.55	3.4	< 0.001	3.4	-	0.0527
5	2.20	13.5	< 0.001	13.5	-	0.2970
6	2.59	12.0	< 0.001	12.0	-	0.3107
7	2.75	2.6	< 0.001	2.6	-	0.0715
8	3.52	2.1	< 0.001	2.1	-	0.0737
Sum		170.9		170.9		2.2947

$E_k = 0.0161$; $N_{E_k} = 0$; $N_p = 1.71$; $n_p = 1.71$
 $E = 2.295$ MeV/dis
 $E = 1.343$ MeV/photon

Where

N = number of nuclear transitions of a given energy per 100 disintegrations
 N_g = number of unconverted gammas per 100 disintegrations
 E_g = energy of gamma photon in Mev
 α = total conversion coefficient
 N_k = number of K x-rays and Auger electrons per 100 disintegrations
 N_p = total number of nuclear transitions per disintegration
 n_p = number of photons per disintegration which escape the atom
 E_k = energy of K x-ray

Some such transitions violate rules governing allowable transition according to quantum theory. (u)

To predict gamma radiation intensities one must know the energies and the abundances of the gamma rays emitted per beta disintegration of a radionuclide. In this respect the term "curie" as used in connection with gamma emitters should be clarified. A curie of beta radioactivity, defined as 3.7×10^{10} disintegrations per second may be associated with emission of more than 3.7×10^{10} photons per second. Gamma transitions are not defined as disintegrations, and the term curie should not be used to express gamma emission rate. (u)

Shielding calculations also require decay scheme data because of the dependence of the absorption coefficient on gamma energy. A high energy gamma ray is less efficiently absorbed than a low energy gamma ray. Comprehensive studies have been made to provide decay scheme data but there is still insufficient information for many radionuclides. A compilation of available data on decay schemes has been used by Miller (1957a, 1957b) to compute decay of gamma radiation from fission products from slow-neutron fission of U^{235} . (u)

10.1.6 NUCLEAR STRUCTURE AND DECAY PROCESSES

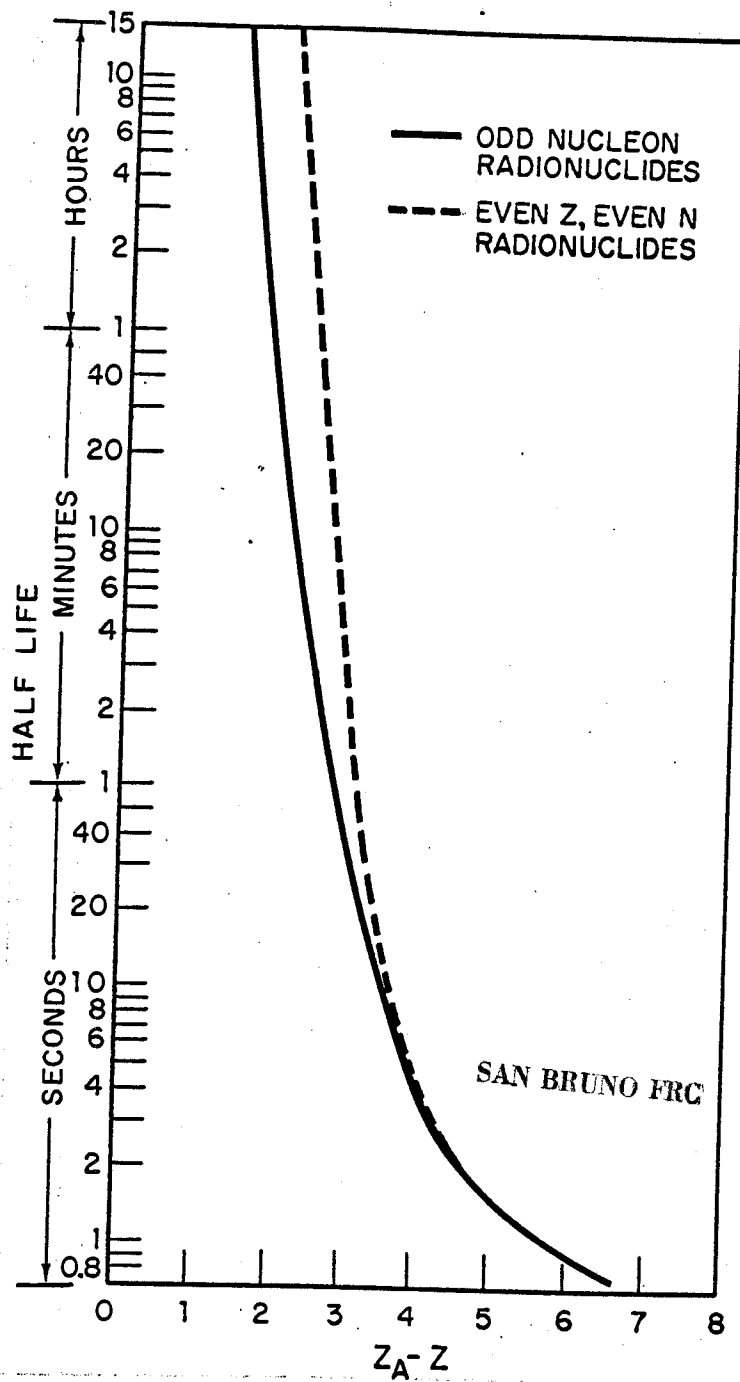
For a discussion of decay processes, it is necessary to consider the composition of the nucleus and the forces acting within it. At one time the nucleus was conceived as a simple assemblage of neutrons and protons. However, now it is thought that the basic particles, or "nucleons," in the nucleus participate in a kind of exchange reaction with entities called π mesons (pions). Pions may be of three types, π^+ , π^- and π^0 , corresponding to +1, 0, or -1 electron charges. The rest mass of π^0 is 264 electron masses, and that of π^+ and π^- is 276 electron masses. The emission of a π^+ by a proton and its capture by a neutron, or conversely, the emission of a π^- by a neutron and its capture by a proton, corresponds to interchange of positions of the two nucleons. A π^0 can be interchanged between nucleons of the same type. This exchange between nucleons accounts for the force holding nuclei together, and it overcomes the large electrostatic repulsive forces between the positively charged nucleons. (u)

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A neutron also becomes involved in these exchange reactions when it is captured by a nucleus, and an amount of energy is liberated which is equal to its binding energy in the nucleus. This excess energy may be lost by gamma emission or by particle emission. In some cases there may be competing modes of decay. Because of the way reactions operate, certain combinations of neutrons and protons result in a greater nuclear binding energy than do others. Nuclides having an odd number of

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Fig. 10.1:7 Half-Lives of Beta-Emitting Radionuclides as a Function of Displacement From the Most Stable Nuclear Charge, Z_A



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Fig. 10.1:8 Contribution of Each Fission-Produced Element to Total Fission Product Activity

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nucleons, for example, tend to be less stable than even-numbered nuclei. The alpha particle, composed of two protons and two neutrons, is especially stable. The rate of decay of nuclei is influenced by such factors, as well as by the decay energy and other factors. The systematics of alpha decay has been worked out for a large number of emitters to the degree that reliable predictions for new radionuclides have been made prior to their discovery. (S)

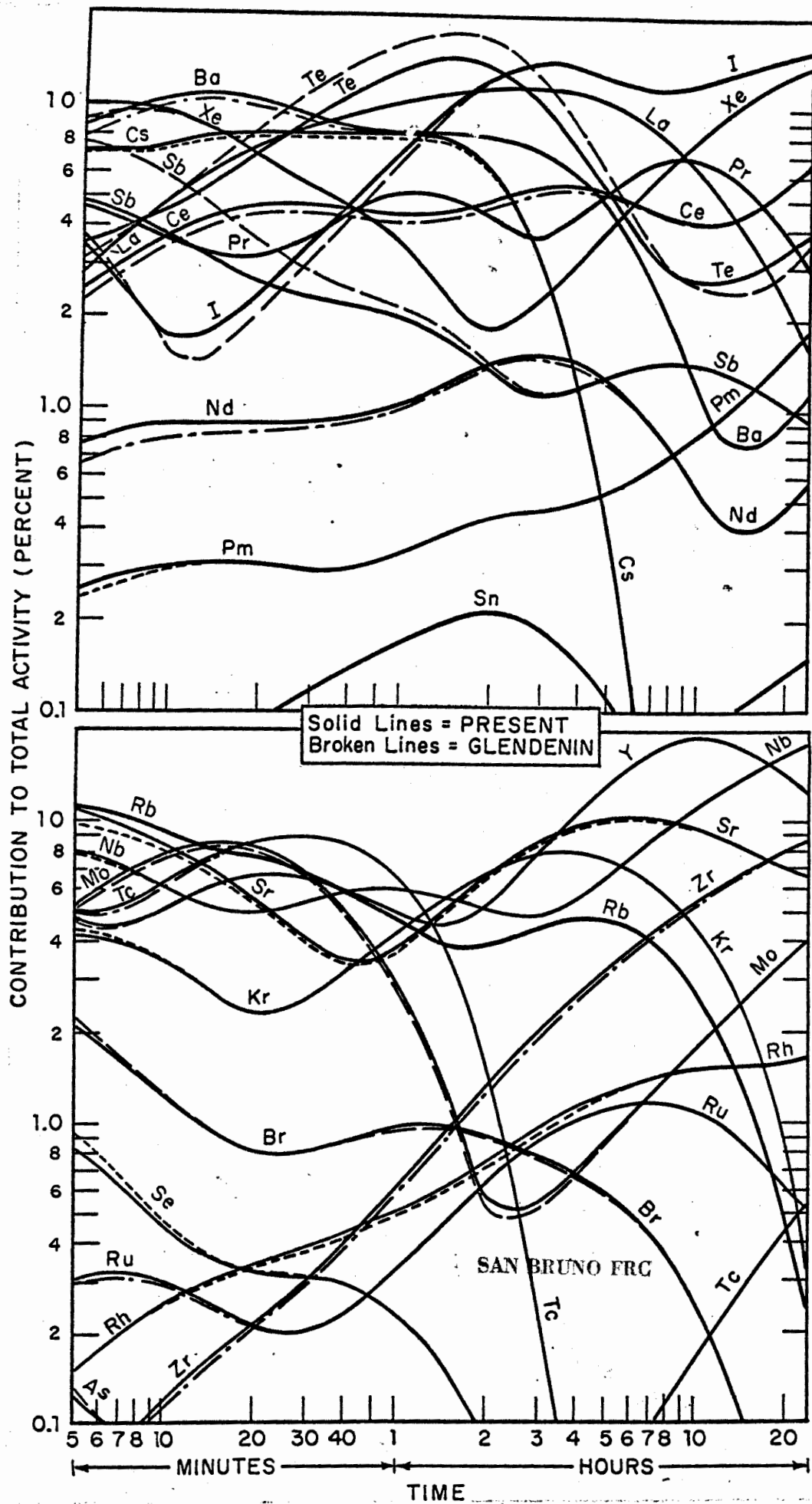
One of the interesting features of beta decay is the continuous spectrum of beta energies observed. The apparent anomaly of identical parent nuclei decaying by emission of beta particles of different energies to identical daughter nuclei once brought into question the conservation of energy and momentum. But this was resolved by ascribing the disappearance of energy to emission of a particle called a neutrino. The neutrino is emitted simultaneously with the beta particle; it has zero charge and zero rest mass. Besides carrying away kinetic energy, it possesses angular momentum and thus conserves both energy and momentum for the beta decay process. Neutrinos have been detected experimentally but exert no observable effect under ordinary circumstances. An expression for the probability of a nuclide undergoing decay by beta emission can be derived from quantum mechanical theory, but this relationship is not a straightforward one. It is sufficient here to note that nuclides farthest from stability have the shortest half-lives (Fig. 10.1:7). (S)

The primary fission products have very short half-lives, but the daughter radionuclides along the mass chain generally are progressively longer-lived. (S)

A useful calculation of the beta activities and abundances of the U^{235} fission products was made by Bolles^{and Ballou} et al. By summing the radioactivities of all the radioisotopes of each fission element at various times, plots can be made of the contribution of each element as a function of time. Figure 10.1:8 illustrates their work, showing the relationships among the various elements from five minutes to 24 hours. (S)

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Similarly the photon emission energy with time can be summed. Miller (1957b) computed the percentages of total photon energy emitted by various radioelements from 0.03 to 10^6 days after fission. Photon data were taken from the nuclear decay schemes of the individual radionuclides as shown in Miller (1957a). Calculations of this type show satisfactory correlation with experimental data, except at very early times where data are lacking, and are useful in interpretation of gamma radiation measurements. The elements that contribute 10 % or more at any time are Kr, Y, Zr, Nb, Rh, Te, I, Cs, Ba, La, Pr, and Sm. (S)



10.2 INDUCED RADIOACTIVITY

Introduction

Although fission products can be regarded as the principal emitters of radioactivity in weapon debris, other materials may also become activated through neutron-induced nuclear reactions and be associated with the debris. Neutrons produced by thermonuclear reactions and excess neutrons produced by fission can undergo different nuclear reactions with device components and with the environment materials, to form radioactive species not found among the fission products. Their contribution to the total radiation varies depending upon the composition of the device, fission yield, nuclear cross-sections of the target nuclei, half-lives of the products, etc. In general, induced activities provide a large fraction of the total activity only in the case of clean thermonuclear weapons; however, significant contributions also are found for other conditions. In this section a discussion of the factors determining these contributions is presented. (u)

The excess of neutrons produced by fission over those required for fission is given by $(\nu-1)$ where ν is the average number of neutrons emitted during fission and has the approximate value 3 and 2.5, for Pu^{239} and U^{235} respectively. If the instantaneous energy release is 177 Mev per fission and a kiloton is defined as 1.0×10^{12} calories, there are 1.46×10^{23} fissions per kiloton. The total neutrons released in fission and available for non-fission absorption processes are, therefore, $1.46 \times 10^{23} (\nu-1)$ per kiloton. (u)

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Ultimately, absorption of the excess neutrons occurs in three modes: (a) captures in the fissionable material which do not result in fission; (b) captures in non-fissionable components of the device such as high explosive products, bomb casing, or tamper; and (c) captures in environmental materials such as seawater and bottom material. The ratio of the number of neutrons absorbed by all non-fission processes within the weapon to the number of fission captures is referred to as the capture-to-fission ratio and is designated by α . The number of neutrons released from the core of a pure fission weapon is thus $1.46 \times 10^{23} (\nu-1-\alpha)W$ where W is the fission yield in KT. α is quite dependent upon weapon design. Values have been reported by Bakal, et al and Rainey in the range 0.05 to 1.5. Boosted and thermonuclear weapons produce larger numbers of neutrons than pure fission devices for equivalent yields. For boosted weapons, of the order of 2.6×10^{23} neutrons per KT are produced and for thermonuclear weapons 1.2 to 2.4×10^{24} neutrons per KT have been reported by Bakal, et al, Cowan, and Klement. Since each excess neutron is ultimately absorbed in a neutron-capture reaction, a greater number of atoms of capture reaction products may be formed than of fission products. However, some of these products are

radioactively stable while others have nuclear decay properties such that their contribution to the total radioactivity of bomb debris is negligible at any time. (S)

The types and quantities of induced activities formed in underwater bursts are discussed and comparisons made with fission product activities in the succeeding sections. (S)

10.2.1 NEUTRON FLUX AND NEUTRON ENERGY SPECTRUM

Values for neutron flux from nuclear devices are extremely high compared with typical nuclear reactor fluxes due to the extremely short period of neutron emission. For the practical estimation of induced activities, neutron emission may be regarded as instantaneous since it occurs within about 5×10^{-5} sec for fission devices and even more rapidly for thermonuclear reactions. A relatively small fraction of fission neutrons is emitted at delayed times by six major groups of fission products whose half-lives for emission vary between 0.15 to 54 sec. According to Bakal, et al, the percent of total neutrons which is delayed is 0.23, 0.69, and 1.47 for Pu²³⁹, U²³⁵, and U²³⁸ respectively. (S)

Calculations of yields of nuclear reactions may be made by use of the following standard formula (thin target):

$$N = N_0 \sigma nvt \quad (10.2:1)$$

where N = Number of transmuted atoms per cm³

N₀ = number of target atoms per cm³

σ = Nuclear cross-section of target atoms in cm²

n = number of neutrons per cm²

v = velocity of neutrons in cm per sec

t = time of irradiation

The product nv has the dimensions of neutrons per cm² sec and is defined as the flux. The product nvt, or total number of neutrons per cm², frequently is determined rather than flux for short neutron exposures. The nvt and the flux decrease with distance due to scattering effects and absorption. (S)

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For calculations of yields of nuclear reactions occurring within the device, internal flux or nvt values must be used. These values are not necessarily uniform throughout a device nor ^{the same} for every device, but according to Cowan, nvt values are of the order of 1×10^{23} neutrons per cm² for fission devices. (S)

Fig. 10.2:1 Typical Fission Neutron Energy Spectrum

The neutrons from fission have an average energy of about 4 Mev, but the neutron spectrum ranges from zero to values well above 4 Mev. A typical fission neutron energy spectrum is shown in Fig. 10.2:1. (S)

Thermonuclear neutrons have characteristically higher energies than fission neutrons, the most prominent value being 14 Mev from the H^2, H^3 reaction. The spectrum for boosted weapons is shifted toward higher energies for this reason. (S)

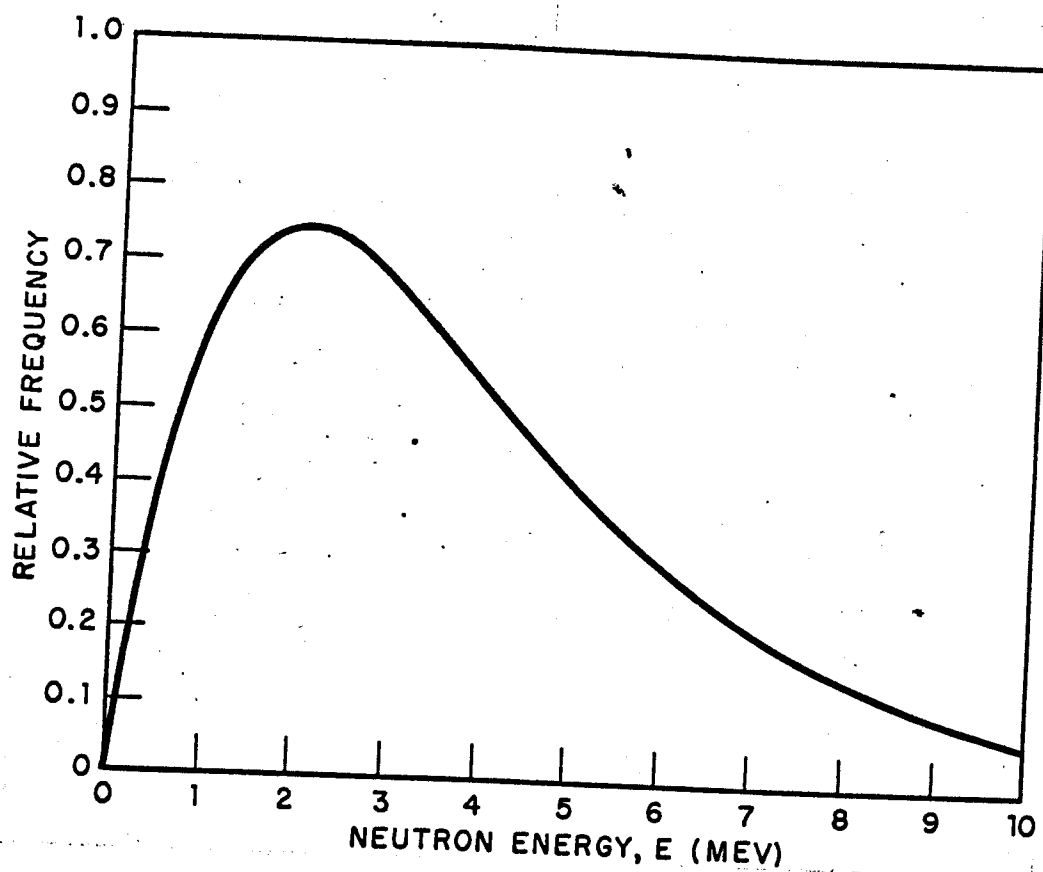
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Rapid degradation of neutron energies occurs after their formation by interactions with the bomb components, especially hydrogenous high explosive. Some neutrons which escape from the device will have been thermalized to bomb temperatures (approximately 1 Kev) but for

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External neutron energy spectra observed in Operation TEAPOT tests are shown in Table 10.2:1 as taken from Bakal, et al.



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Table 10.2:1

Neutron Energy Spectra of Unboosted and Boosted Fission Weapons

Total Flux (%)

Neutron Energy
Range

4 kev - 0.75 Mev
0.75 - 3 Mev
> 3 Mev

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In the case of an underwater burst, neutrons escaping from the device encounter a hydrogenous medium, water, which results in complete thermalization of all neutrons to ambient temperature within times much less than a millisecond. (S)

10.2.2 MODIFICATION OF NEUTRON FLUX IN WATER ENVIRONMENT

Two important effects occur when neutrons enter a water environment, thermalization and absorption. The average free-flight distance traveled by a neutron in slowing down is described by Glasstone, et al (1952) as 14.05 cm, whereas the average free-flight distance traveled by a thermal neutron from point of thermalization to point of capture is 4.157 cm. Thus, it may be seen that both effects occur relatively close to the point of neutron generation. Therefore the important consequence of these observations is twofold:

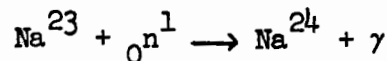
(a) All induced radioactivities produced external to the weapon are completely enclosed within the initial steam bubble and mixed with the weapon debris. SAN BRUNO FRC

(b) No escape of neutrons to the atmosphere occurs except for weapons detonated essentially at the surface; therefore underwater bursts are not accompanied by nitrogen-capture gamma rays. (S)

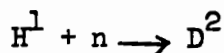
10.2.3 NEUTRON ACTIVATION REACTIONS

The principal type of nuclear reaction occurring externally to the device is radiative capture or n, γ reaction in which a thermal neutron

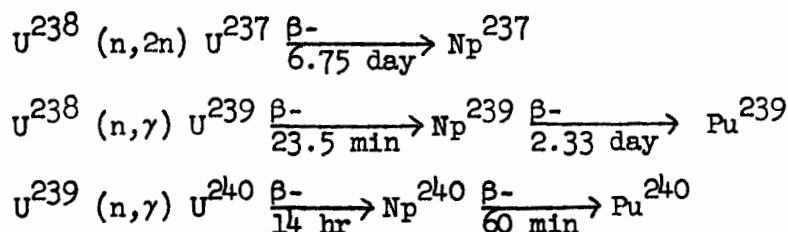
is absorbed and a gamma ray is emitted by the product nucleus, for example:



The products of some reactions are not radioactive, viz., neutron capture by hydrogen



According to Rainey this accounts for absorption of some 67 % of neutrons emitted from the device. At higher neutron energies other types of reactions having a higher energy threshold also occur, e.g., n,2n, n,p, and n, α . Owing to the rapid thermalization of fission neutrons these reactions occur mainly within the fissile material. In weapons containing appreciable amounts of U^{238} , n, γ and n,2n reactions give rise to the most significant of the induced activities. These include the following:



The reaction producing U^{240} is termed second order in that it comprises two successive neutron captures starting with U^{238} . Exact calculations of yields of induced activities would have to consider the U^{238} and its variation with distance from the device, the neutron energy distribution, and the variation of cross-section with neutron energy for all target elements. However, for estimation of yields from thermal neutron capture, it may be assumed that all neutrons emitted from a device undergo n, γ reactions, and that the relative number of each type of product nucleus is proportional to the product $N_i \sigma_i$

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where N_i = number of atoms of the i th isotope per unit volume

σ_i = cross section of the i th isotope for thermal neutron capture

There will be $(v-1-\alpha)W$ total product nuclei, and the number of atoms of the i th isotope N_i is given by

Integrated Neutron Flux

$$N_1 = \frac{n_i \sigma_i}{\epsilon n_1 \sigma_1} (\nu - 1 - \epsilon) W \quad (10.2:2)$$

10.2.4 PRIMARY RADIONUCLIDES PRODUCED BY NEUTRON ACTIVATION

The radioactivity of the induced products can be calculated by use of the equation

$$\frac{-d N_1}{d t} = \lambda_1 N_1 \quad (10.2:3)$$

where

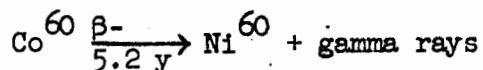
λ_1 = nuclear decay constant

and is related to the half life $t_{1/2}$ by

$$t_{1/2} = \frac{\ln 2}{\lambda} = \frac{0.69315}{\lambda} \quad (10.2:4)$$

The principal seawater constituents are shown in Table 10.2:2, and results obtained by Rainey for the most important neutron capture products are given in Table 10.2:3. The significance of these amounts of radioactivity compared with fission product activity is discussed in 10.2.5.

Thermonuclear detonations ~~would~~ produce more induced activity relative to fission products. This effect could be enhanced by the "salting" of weapons, that is, the introduction into the device of inert materials selected to give specific induced radioactivities having desired half-life or gamma-emission properties. For example, the following reactions using natural cobalt might be used:



~~Generally speaking, such deliberate production of radioactive materials to achieve military objectives is considered to have limited applicability. (S)~~

A very important source of early gamma-radiation from above-surface detonations arises from capture of neutrons by atmospheric

Table 10.2:2

Principal Constituents of Seawater

ANALYST	Gms per liter at 20°C (sp.gr. 1.025)
sodium	11.1
magnesium	1.33
calcium	0.42
potassium	0.39
strontium	0.01
chloride	19.8
sulfate (as SO_4)	2.76
bromide	0.066
boric acid (as H_3BO_3)	0.026
carbon	approx. 0.026
nitrogen	0.013
hydrogen (as water)	111
oxygen (as water)	878

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Table 10.2:3

Principal Products of Thermal Neutron Capture in
Seawater*

Target Isotope	Atoms per liter x 10 ⁻²³	Thermal Cross Sections (cm ² x 10 ²⁴)	n _i σ _i (atom-cm ²)	$\frac{n_i \sigma_i}{en_i \sigma_i}$	Product Isotope	t _{1/2}	$\lambda_i N_i$ per KT Disintegrations per sec
H ¹	686.1	0.330	22.64	0.672	D ²	Stable	None
Na ²³	2.836	0.560	0.159	0.00472	Na ²⁴	15.0 hr	2.01 x 10 ¹⁶
Cl ³⁷	.807	0.560	0.0434	0.0013	Cl ³⁸	37.3 min	1.32 x 10 ¹⁷
Cl ³⁵	2.49	44	10.90	0.324	Cl ³⁶	3.1 x 10 ⁵ y	5.14 x 10 ⁹

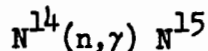
$$en_i \sigma_i = 33.74 \text{ atom-cm}^2\text{-l}^{-1}$$

(1) Based on $(\nu-1-\epsilon) = 1.5$

*Oxygen and boron capture a few percent of the thermal neutrons but result in no significant product radioactivities; hence, have been ignored in this calculation. (8)

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nitrogen and nitrogen in the high explosive component of weapons by the reaction



N^{15} is not radioactive, but the capture gamma radiation is quite energetic. Several gamma rays are emitted having energies reported by Kinsey from 4.5 to 10.8 Mev. Similarly, according to Wapstra the capture of a neutron by hydrogen results in emission of 2.2 Mev gammas. These emissions are of interest only in surface or near-surface detonations. At the surface, activation of both atmospheric nitrogen and hydrogen occurs. At increasing depth, even a thin layer* of water over the fireball prevents escape of neutrons to the atmosphere except for delayed neutrons. From Russell, et al, it can be seen that dose rates delivered by capture gammas from delayed neutron reactions appear to be less than a percent of those delivered by fission product gammas. For thickness of water over the fireball of ~ 10 to 100 ft, attenuation of capture gammas by water reduces the surface radiation level to insignificance. The contribution of neutron capture gammas to very early gamma radiation fields for near-surface depths has not been estimated, but compared with the gamma fields from fission products, it is not considered to add significantly to the dose delivered in the range where radiation effects are predominantly of interest. (S)

10.2.5 CONTRIBUTION OF INDUCED ACTIVITY TO FISSION PRODUCT DEBRIS

The activity with time of the two principal activities, Na^{24} and Cl^{38} , induced in seawater may be calculated from the standard equation:

$$I = I_0 e^{-\lambda t} \quad (10.2:5)$$

where I = activity at time t in disintegrations per second
 I_0 = activity at time zero in disintegrations per second
 λ = radioactive decay constant

The fission product decay curve may be approximated by

$$I = I_0 t^{-1.2} \text{ SAN BRUNO FRC } (10.2:6)$$

Rainey's computations suggest the ratio of induced activity to fission product activity reaches a maximum at 26 hrs. It has the value 0.027

*See Section 10.2.3

at this time and is attributable almost entirely to Na^{24} . Thus, for pure fission weapons a maximum of a few percent of the activity arises from induced products in seawater.*

The contribution of heavy-element-induced activities to total activities also has been calculated by Dolan and Knapp. These results have been expressed as gamma dose or dose rate with time at 3 feet above a smooth plane on which the products of 1 KT are assumed to be spread uniformly. ~~Reference to Figs. 10.2.1, 10.2.2 (Dolan's Figs. 4 and 5), and Fig. 10.2.3 (Dolan's Fig. 3) shows the heavy element contribution to dose rate at three days, for example.~~

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In summary, it can be stated that fission weapons having little U^{238} will have, at most, a few percent contribution of induced activities to total activity; those containing appreciable U^{238} may have induced activities contributing upwards of 10 %; whereas only for clean thermonuclear devices will induced contributions substantially exceed 10-25 % of the total.

10.3 FRACTIONATION

In previous sections of this chapter, a description was given of the sources of radioactivity from underwater nuclear detonations, and of how the composition of the source changed with time due to radioactive decay of the constituents. Variations in the original source composition were seen to be a result of the nuclear character of the detonation, including nuclear reactions with environmental materials. Other processes causing variation in the composition of radioactive debris are a consequence of the chemical properties of the radioactive constituents. The net result of these processes is that samples of the debris and their associated radioactivity are non-representative of the debris taken as a whole, having been either relatively enriched or depleted with respect to some constituents. The result of such processes is referred to as fractionation. The simplest example of its operation is the condensation of refractory elements out of gaseous mixtures. As

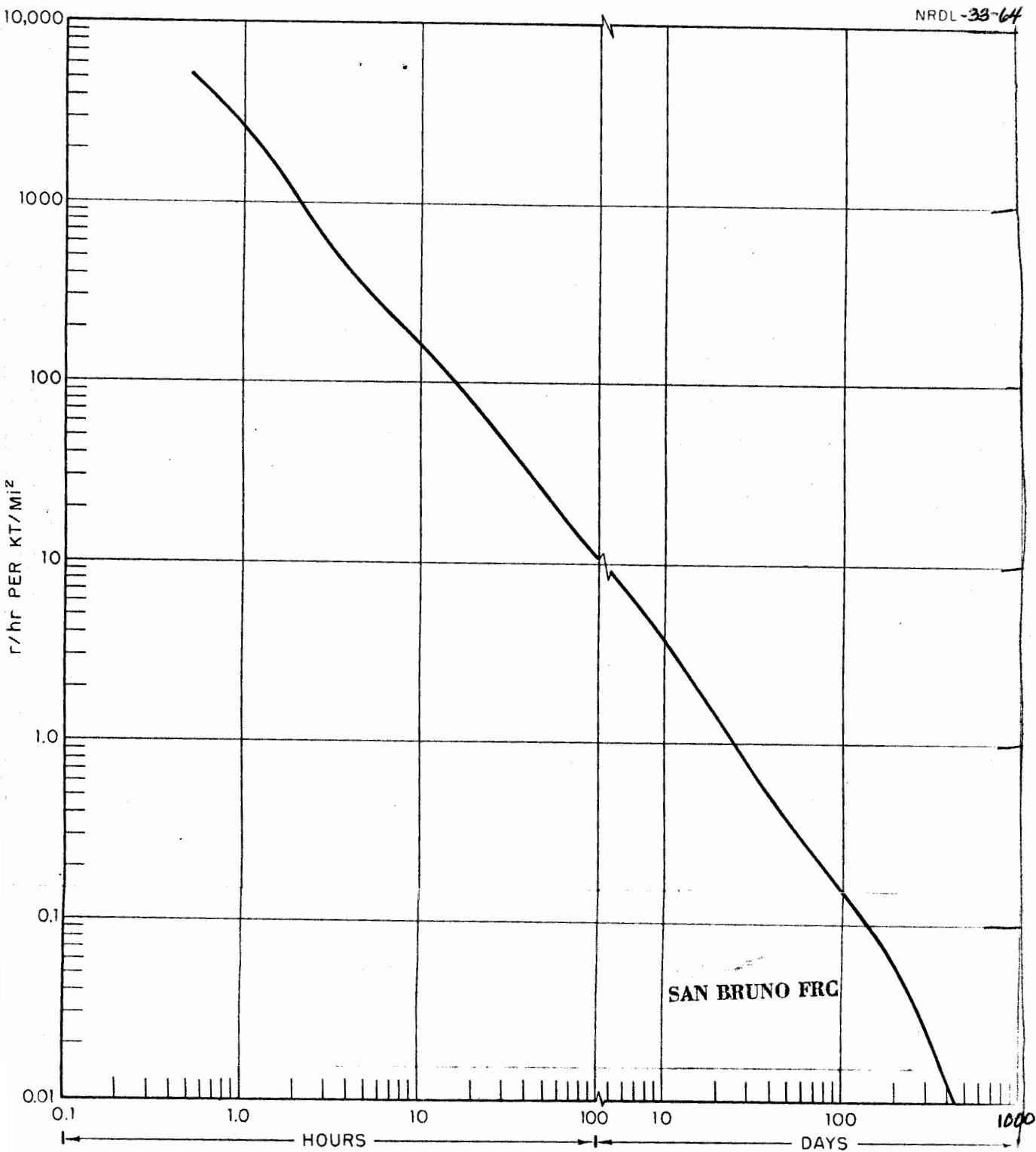
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* These calculations refer to beta disintegrations. The relative contributions to gamma radiation may be assumed to be similar since both Na^{24} and Cl^{38} emit energetic gamma rays.

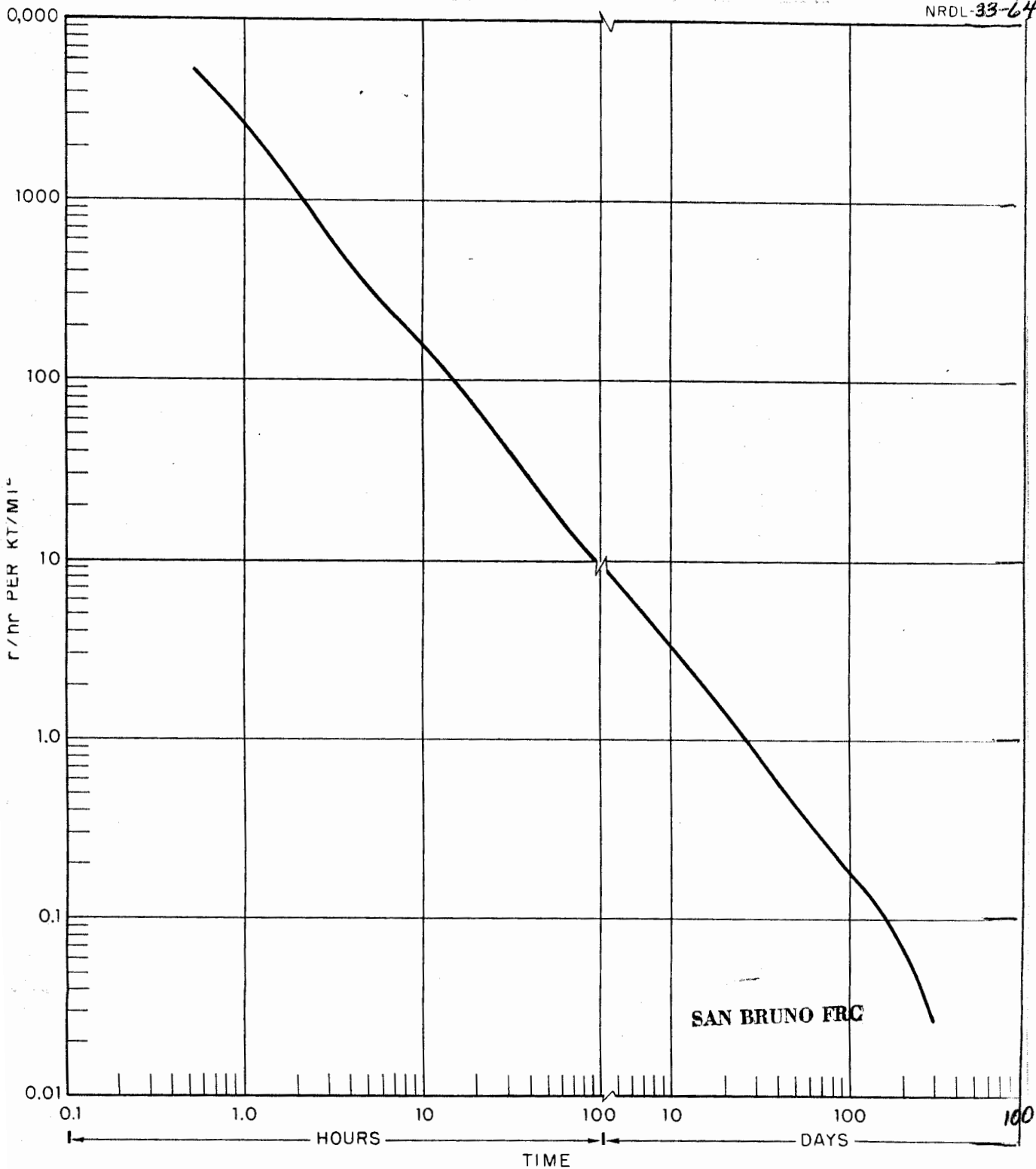
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Other processes causing variation in the composition of radionuclide debris are a consequence of the physical and biological environment.

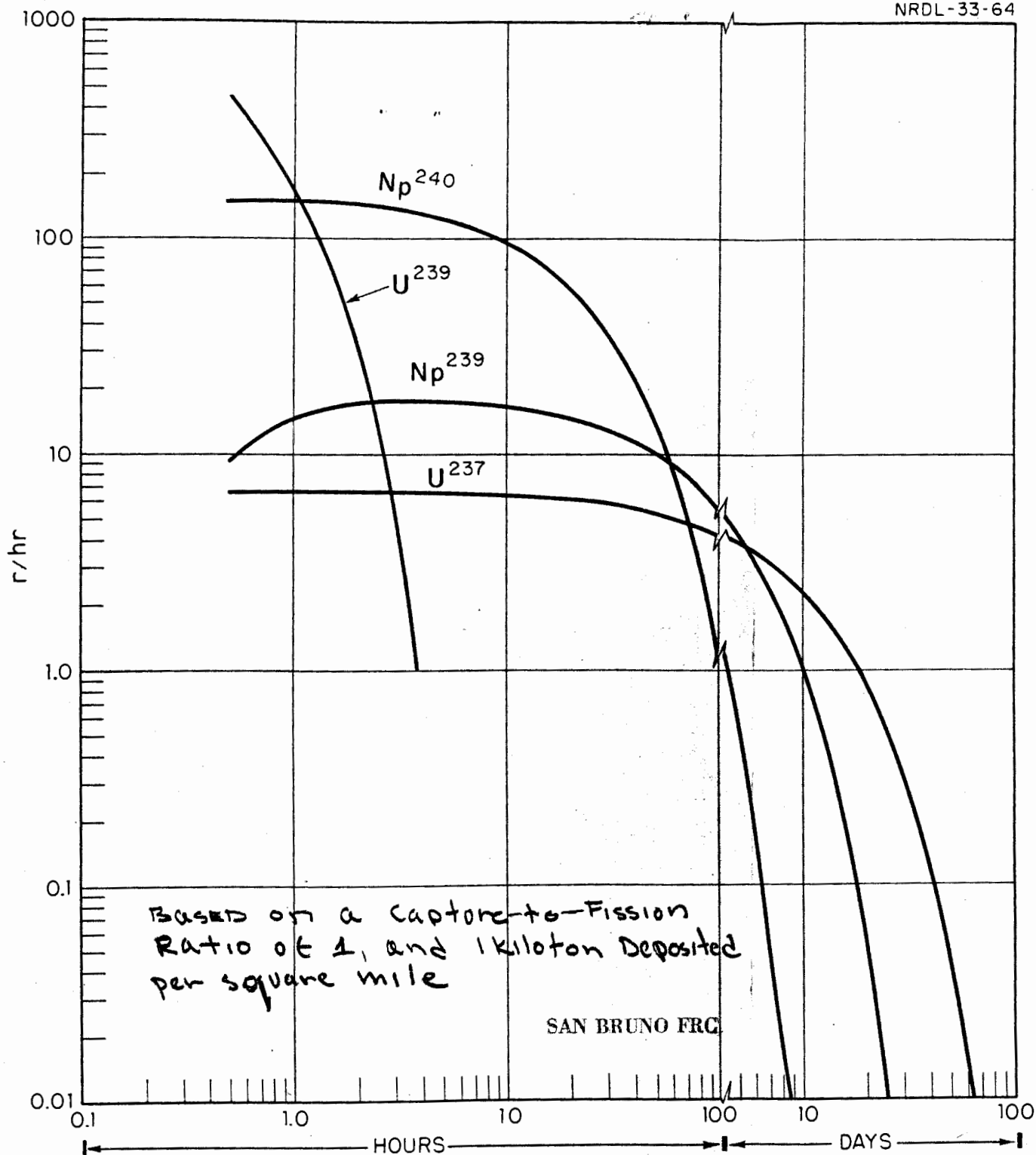
Reference to Figs 10.2.12, 10.2.13 and 10.2.14
(Dolan's Figs 4, 5 and 3 respectively) and
the product-to-fission ratio for each product



10.2:2 Fig. 10.2:2 Typical Dose Rate Decay curve For A megaton YIELD weapon.



10.2.3 Fig. 10.2.3 Typical Dose Rate Decay curve for a
KILOTON YIELD WEAPON
32



10.2:4 Fig. 10.2:4 DOSE RATE TIME DECAY CURVES FOR SOME INDUCED 32 ACTIVITIES

a consequence of fractionation, for example, deviations from calculated values have been observed in decay rates of radiation fields produced by bomb debris. (S)

10.3.1 DEFINITION

burst The term "fractionation" is defined by Freiling (1961) as any alteration of radionuclide composition occurring between the time of ~~detonation~~ and the time of radiochemical analysis which causes a debris sample to be non-representative of the ~~detonation~~ products taken as a whole. The alteration observed may have taken place in various stages, and it is helpful to classify these according to the types of processes involved in their chronological order. (S)

burst "Natural fractionation" begins with the condensation of radioactive and inert material from the fireball. Some radioelements are preferentially taken up in a condensed phase which probably forms within seconds after ~~detonation~~, perhaps prior to the time the gas bubble reaches the surface for deep detonations. Subsequent contact of the debris with condensed water droplets or dispersed seawater (throwout and base surge) provides further opportunity for separation of non-volatile radionuclides from volatile ones. The fractionation taking place through these processes is called "primary fractionation." (S)

Further fractionation may then occur through contact of the debris with the surroundings. Soluble radioelements will dissolve in seawater whereas the insoluble will remain in a particulate or colloidal state. Solid particles may adhere to surfaces, and dissolved radioelements may become adsorbed. Fractionation occurring by processes such as these is called "secondary fractionation." (S)

"Artifactitious fractionation" can be induced by sample collections that result in biased samples, by incomplete removal of debris from sampling apparatus, and by faulty analytical procedures. It has also been observed that fractionation results from certain radiological recovery operations such as decontamination methods. (S)

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A common but poor technique for assessing the extent of fractionation is to compare radionuclide abundance ratios in samples of bomb debris with ratios for thermal fission of U^{235} or with ratios for other detonations. It does not follow that observed differences necessarily indicate that fractionation has, in fact, occurred, but may merely reflect the true composition of the detonation products for that type of device. Taking the debris from a weapon as a whole, the relative yields among fission products from each type of device depend upon the nature of the fissioning nucleus and upon the energy spectrum

of the incident neutrons. Direct evidence of the occurrence of fractionation is provided by variations in composition among samples of debris from the same detonation, or among samples of debris from detonation of identical weapons under different environmental conditions. (u)

10.3.2 Measurement Techniques

Freiling (1961) has cited some interesting correlations among the various nuclide ratios which aid in the determination of fractionation. They are illustrated with the following relationships.

Let $F_i = A_i/Y_i$ 10.3:2

where F_i = total number of fissions which occur

A_i = total number of atoms of radionuclide i

Y_i = atoms of radionuclide i produced per fission

In this case Y_i relates to total fission and radionuclide yields for a particular device rather than to yields for thermal neutron fission of U^{235} and thus eliminates nuclear physical factors which are irrelevant for consideration of fractionation effects. All values of F_i should be equal. If, however, only a sample of the products is available for analysis, and the number atoms A_i are determined by radiochemical analysis, values F_i may be similarly calculated as

$f_i = A_i/Y_i$ 10.3:3

but agreement will be obtained only among those radionuclides which have not fractionated from one another, and variance of f_i values will be indicative of fractionation. With Zr^{95} and Sr^{89} chosen as reference radionuclides, a further relationship is defined.

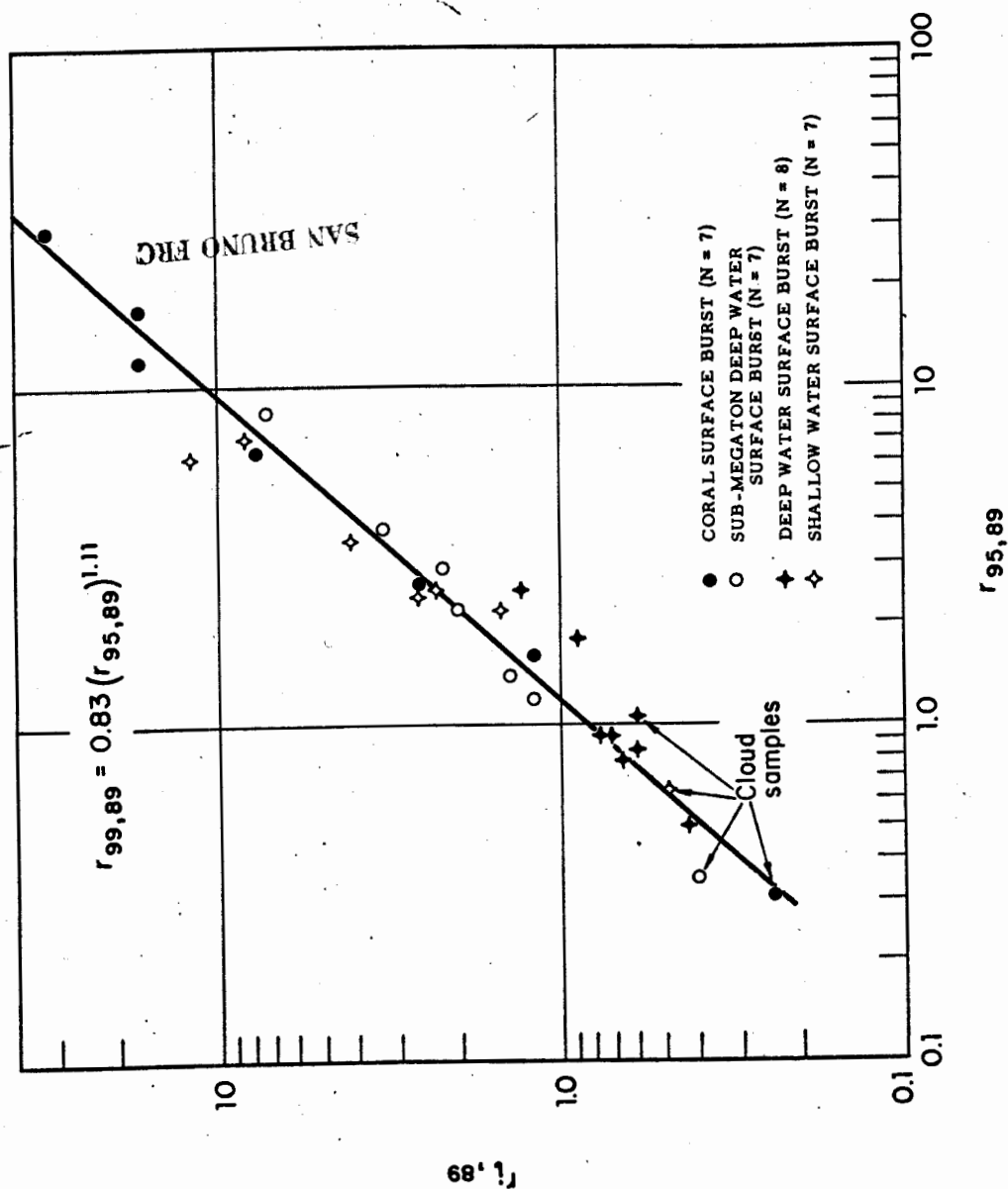
SAN BRUNO FRG

$r_{95,89} = f_{95}/f_{89}$ 10.3:4

to positive values greater than unity for other isotopes. However the values beyond 0 and 1 were not statistically significant. An interesting empirical correlation also was found to exist between the slope and the volatility of the precursors, expressed as the square root of the fraction F_R of a decay chain existing in a refractory form at a given time. This relationship at $t = 35$ sec is illustrated in Fig. 10.3:2. (u)

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Fig. 10.3:1 Logarithmic Fractionation Correlation for Molybdenum-99

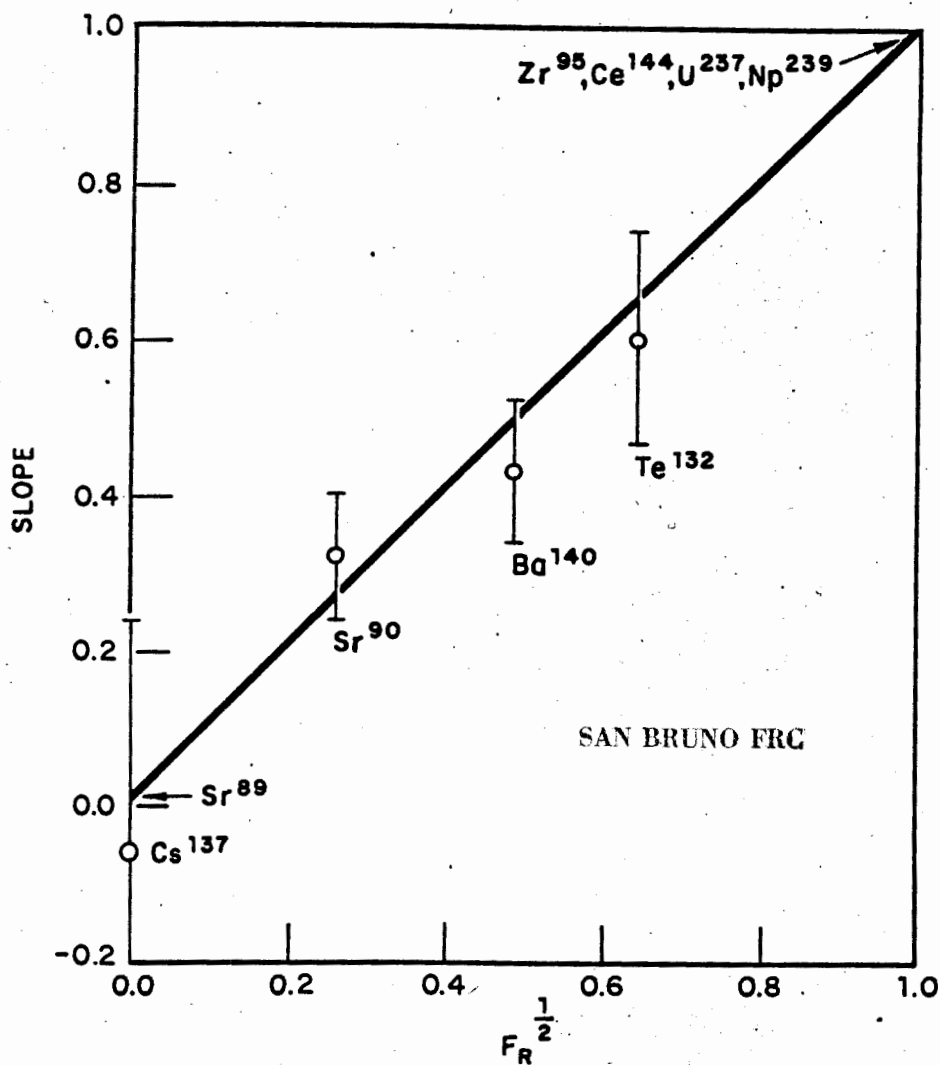


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Fig. 10.3:2 Dependence of Correlation Slope on Volatility of the Precursor

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4"4
(24 ± 90)

Red. to 3 7/8"

From the above discussion, it is apparent that empirical relationships exist among experimentally observed fractionation effects which may be related to the chemical properties of the mass chain at the time of condensation. Although much more data is needed, these relationships provide a valuable empirical basis for prediction of unfractionated composition of bomb debris, and for estimation of the fractionation behavior in future detonations. A theoretical basis underlying these effects has recently been published (Freiling 1963).

10.3.3 CAUSES OF FRACTIONATION

The processes which give rise to fractionation begin at early times in the fireball and continue throughout the sequence of dynamic surface phenomena, e.g., base surge formation. Theoretical treatments of the subject given by Magee, Miller and Caputi have contributed to the development of a quantitative description of the phenomenon, but has not yet provided a generally applicable theory. Nevertheless, it is thought that the main factors giving rise to this effect are fairly well understood, at least in qualitative terms.

The initiation of nucleation within the fireball can be anticipated by the time the temperature has dropped below the boiling points of device and seawater constituents. For land-surface bursts this should occur below 2800°K , the boiling points of CaO and MgO , and the approximate point where iron oxide begins to condense according to Freiling, et al (1962) and Adams et al. In seawater a distribution of fission product elements between vapor and liquid phases can be thought of in accordance with laws of solution and the partial pressures of the vapors of the various fission elements over the mixed oxide melts. Additional phases form when the temperature has dropped below the boiling point of NaCl (1686°K) and the sublimation point of Na_2O (1550°K). A qualitative idea of the relative tendency of fission product elements to condense at this stage can be obtained from Table 10.3:1 citing Miller's (1950) work which shows the partial pressures of several elements over their own oxides at 1673°K . At this temperature La , Ce and Zr would have condensed to the solid state and would be mixed with solid oxides of the inert constituents. With further cooling, the other elements also condense but become associated with the lower melting point constituents such as NaCl , and eventually water. Obviously, the elements Kr and Xe remain in the gaseous state, but their decay products enter into the condensation reactions.

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The half-lives of a number of the important rare gas nuclides are comparable with the times of cooling of the fireball, of the order of one second to a minute or less depending upon yield. Some rare gas nuclides have longer half-lives, comparable with the times over which

Table 10.3:1

Partial Pressures of Some of the More Abundant Fission
Products Over Their Own Oxides at 1673°K

<u>Element</u>	<u>Partial Pressure Over Oxide (atmos)</u>
Rb	1.2 (1)
Cs	3.2 (1)
Sr	1.3×10^{-8} (s)
Ba	5.0×10^{-7} (s)
Ia	2×10^{-15} (s)
Ce	8×10^{-13} (s)
Sb	0.34 (1)
Zr	2×10^{-14} (s)
Nb	1.6×10^{-7} (s)
Mo	3.6 (1)
Te	4.2 (1)

s for solid
l for liquid

secondary fractionation processes take place. The operation of these processes for an underwater burst may be visualized as follows: water which condenses either prior to or following interaction of the steam bubble with the surface contains relatively small amounts of dissolved rare gases, but it contains the majority of the non-gas fission product nuclides. As further contact between gaseous and liquid water phases is maintained within the subsiding above surface plumes and precipitation from the base surge, further effective scavenging of these non-gas nuclides may be assumed. Therefore within the cloud of the base surge aerosol, the fission product rare gases are enriched with respect to the other fission products. However upon decay to a nonvolatile daughter element, a nuclide is rapidly scavenged owing to the large surface area of the liquid aerosol. Therefore, the base surge droplets become increasingly enriched with time in the decay products of the gaseous radionuclides as experimentally indicated by Evans et al. As a result of this process, enrichments of Sr^{89} in base surge by factors as high as 2000 have been reported for samples from Shot Umbrella, Operation Hard-tack. After the radioelements have been scavenged by contact with water, secondary fractionation effects also occur, as described earlier. ~~///~~

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Considerably more information on the mechanisms of fractionation in an underwater detonation certainly is needed before the above hypothesis can be substantiated in detail. However radiochemical and

radioactive decay data obtained during field experiments are in qualitative agreement with the mechanism described. (S)

10.3.4 IMPORTANCE IN DEFINING RADIATION FIELDS

The existence of fractionation has, of course, important consequences for diagnostic measurements, determination of fission yield from bomb debris samples, etc. For high yield surface shots, it also accounts for the relative enrichment of $\text{Sr}^{89,90}$, I^{131} and Cs^{137} in world-wide fallout over that in local fallout. (S)

To the extent that fractionated rather than representative bomb debris is either deposited or transported following underwater detonations, the resulting radiation fields may be expected to deviate from those produced by a nonfractionated radiation source both in intensity and in decay characteristics. Miller (1950) for example has calculated theoretical disintegration rates and air ionization rate for fractionated fission products condensed within particles of an idealized carrier material. The results are compared with the ionization rate calculated for normal products of thermal fission of U^{235} in Fig. 10.3:3. (S)

Although the applicability of Miller's theory to underwater conditions remains to be established, the curves serve to show qualitatively the decrease in radiation field which can result from depletion of the volatile fission nuclides in fallout. Experimental evidence for the effects of relative enrichment and depletion was obtained from two underwater nuclear detonations, Wahoo and Umbrella, where base surge samples analyzed by Schell, et al and Evans, et al showed variations in rates of gamma ionization decay. Water samples from underwater shots would be expected to show relatively less fractionation, and this is confirmed by results obtained by Ballou at Operation Wigwam. (S)

Much more information is still needed before a proper assessment of fractionation can be made for underwater shots. Data are needed on the radiation properties of the short-lived fission products, independent fission yields, and the way in which these yields vary with fissile material and neutron energy. The basic chemistry of mixed oxide systems, including thermodynamic and reaction rate data for these systems are also areas needing study. (S)

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Finally, further development of theoretical aspects of the fractionation process are necessary to permit evaluations for a range of yields and detonation conditions. (S)

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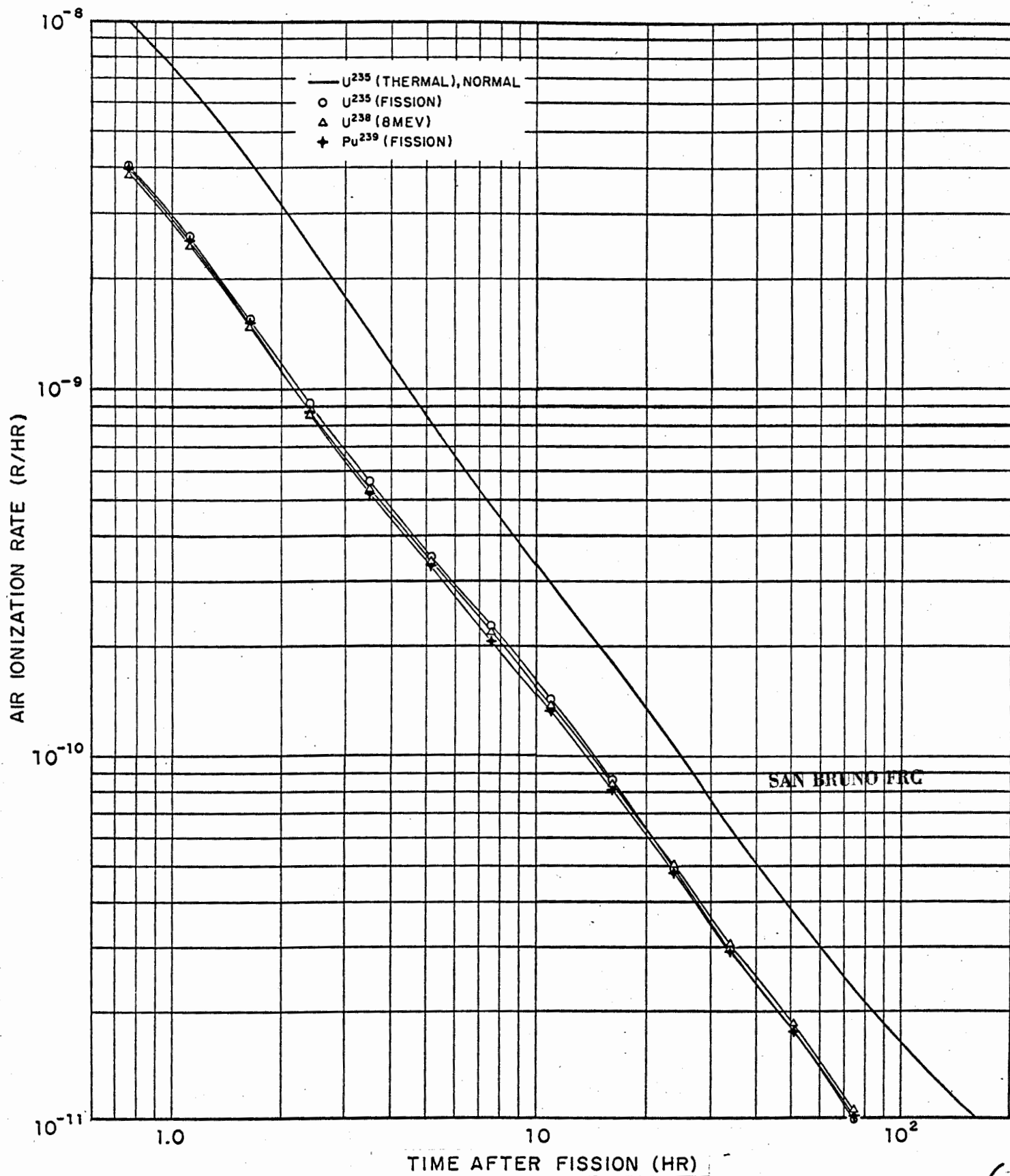
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Fig. 10.3:3 Air Ionization Rate at 3 ft Above a Smooth Infinite Plane Uniformly Contaminated With Fractional Fission Products From 10^4 Fissions/sq ft in Melted Fallout Particles From a 2.3 MT Yield Surface Detonation. Rates for normal fission products from thermal neutron fission of U^{235} are given for comparison.

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10.4 MEASUREMENT OF IONIZING RADIATION

Generically, the term "radiation" indicates the flow of energy through space. It is quite distinct from radioactivity which refers to the spontaneous decay of certain types of unstable nuclei. Ionizing radiation primarily differs from other forms such as visible light and radiowaves in that each photon or particle has sufficient energy to strip electrons from atoms which it encounters, i.e., to ionize them. The resulting ions are then in a highly energetic state and their reactions provide the ultimate effects observed from ionizing radiation. (N)

Those radiations associated with radioactive materials are normally limited to alpha, beta, gamma and occasionally neutron radiation. These are described earlier in this chapter and are summarized briefly below. (N)

Beta radiation consists of streams of electrons of greater than thermal energy passing through space or matter. Beta particles are identical in every respect to electrons, but the term has been applied historically only to those electrons emitted from the nucleus of an atom. (N)

Alpha radiation consists of streams of helium atoms which have been stripped of both orbital electrons. Each particle has an atomic mass of four units and a positive charge of two units. Because of its mass and charge an alpha particle reacts very strongly in passing close to other atoms. It, therefore, has a very limited range of only a few centimeters in air, and can be stopped by a thin sheet of paper. (N)

As mentioned earlier gamma rays are defined as electromagnetic radiation produced in nuclear reactions as compared with X rays which are formed in the excitation or removal of orbital electrons by the deceleration of electrons. Gamma and X rays consist of photons each with a specific energy, and their interaction with matter occurs by three mechanisms: (1) the photoelectric effect, (2) Compton scattering, and (3) pair production. These are illustrated in Fig. 10.4:1. Removal of photons from a beam by absorption is proportional to the intensity i of the beam, that is, $di/dx = -\mu i$. In the integrated form $i = i_0 e^{-\mu x}$ where i_0 and i are, respectively, beam intensities before and after passage through an absorber having a thickness x and absorption coefficient μ . It is important to note that it is theoretically impossible to remove all photons from a beam of gamma rays. (N) SAN BRUNO FRC

Neutrons have no charge, an atomic mass unit (amu) one fourth that of an alpha particle and approximately 1,840 times that of an electron. Because of this lack of charge, the neutron does not interact directly

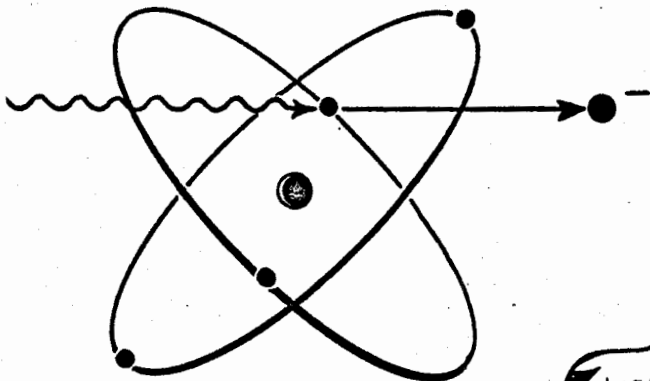
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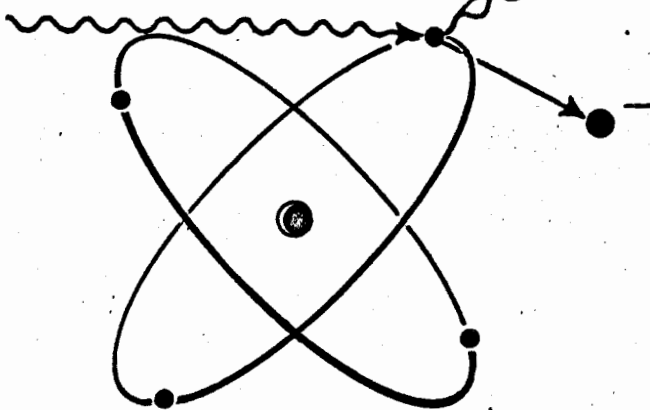
Fig. 10.4:1 Mechanism of Interaction of Gamma Rays with Matter

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PHOTOELECTRIC PROCESS

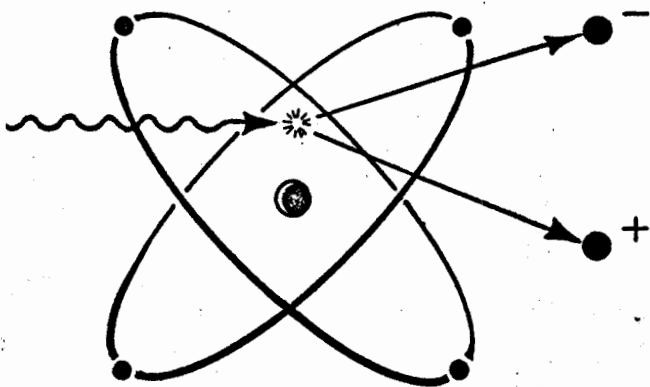
- || γ RAY COMPLETELY ABSORBED
- || ELECTRON EJECTED WITH γ RAY'S ENERGY MINUS BINDING ENERGY



COMPTON RECOIL PROCESS

- || γ RAY OF LOWER ENERGY PROCEEDS IN NEW DIRECTION
- || ELECTRON IS EJECTED WITH THE ENERGY DIFFERENCE

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PAIR PRODUCTION PROCESS

- || γ RAY ANNIHILATED
- || ELECTRON AND POSITRON CREATED AND SHARE γ RAY'S ENERGY MINUS 1.02 MEV

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with an atom to form ion pairs. However, as described in 10.1.4 they can act indirectly either by elastic or inelastic scattering and by neutron capture which results in secondary ionization. (S)

Although alpha, beta, gamma and neutron radiations are the principal ones encountered, there are numerous other forms of ionizing radiation. These include high energy protons (nucleus of a hydrogen atom having unit positive charge, and a mass equivalent to that of a neutron), fission fragments (consisting of high energy nuclear fragments resulting from nuclear fission), cosmic radiation (consisting of atoms of elements up to about oxygen and having an extremely high kinetic energy), and numerous types of mesons. (S)

10.4.1 RADIATION FIELDS, DOSE, AND DOSE UNITS

Ionizing radiation when passing through matter, may deposit a portion of its energy in the medium. Such a radiation flux can be produced by a radioactive source, by fission, by thermonuclear reactions, or particle accelerators. The energy eventually appears in the form of ion pairs which are very reactive chemically. Even though the total energy deposited may be very small compared with that absorbed from normal sunlight, for example, the capacity for production of damaging effects is greatly enhanced by the chemical reactivity. (S)

The quantitative measure of the interaction between radiation and matter is expressed in terms of the energy deposited in the material through which the beam passes, and is referred to as the radiation dose. Evaluation and measurement of this dose is referred to as radiation dosimetry. (S)

Absorption of radiation in air provides the basic unit of radiation dosimetry - the roentgen. Specifically, the definition in physical terms of a dose of one roentgen is an exposure to X or gamma radiation such that the associated corpuscular emission (ionization) per 0.01293 grams of air (1 cc of air at STP) produces ions carrying one electrostatic unit of electricity of either sign. The roentgen, thus defined on the basis of its production of ions in air, is not ideally suited for expression of the dose deposited in materials other than air. If living tissue is exposed to a radiation field of such strength that a dose of one roentgen would have been deposited in an equal volume of air, it is found that approximately 93 ergs of energy are deposited per gram of tissue. (S)

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For practical purposes, the trend has been towards use of a unit based upon the amount of radiant energy deposited in the medium rather than upon the number of ion pairs produced. One such unit is the rad.

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The rad is defined as the deposition of one hundred ergs per gram of tissue in any medium independent of the type of radiation or the medium. This unit provides a useful quantitative measure of deposited dose and is rapidly replacing the roentgen as the preferred unit in dosimetry. It is approximately equivalent to the roentgen for most cases; however, it has a much wider range of use and is suitable for all types of radiation. The roentgen is strictly applicable only to gamma radiation in air and only over a limited portion of the energy spectrum (0.05 to about 2 Mev) beyond which it is not suitable. (u)

Another unit used is the rep. This is defined as the quantity of ionizing radiation which produces, per gram of tissue, an amount of ionization from gamma radiation equivalent to a dose of one roentgen. At one time, the rep was widely accepted as a unit of absorbed dose representing from 93 to 97 ergs per gram of soft animal tissue. Although the rep is still used in some studies, it is quite rapidly being replaced by the more precisely defined rad unit. (u)

In biological exposures the damaging effect of the radiation upon living tissue is of primary interest. Different types of radiation exhibit different degrees of biological effect. In order to express numerically this difference, the concept of relative biological effectiveness (RBE) has been devised. The RBE is a number ranging from 0.5 up to 20 which indicates the relative damaging effect of a particular type of radiation. An extension of this concept gives rise to the rem (roentgen equivalent man), the unit used to express human biological dose as a result of exposure to different types of ionizing radiation. The dose in rem is equal to the absorbed dose in rads times the RBE for the particular type of radiation being absorbed. (u)

10.4.2 SUMMARY OF THE BASIC CONCEPTS OF RADIATION MEASUREMENT

The measurement of any physical phenomenon depends upon some correlation between the signal obtained and the quantity being measured. For radiation dosimetry the quantity being measured is ionization expressing the energy absorbed. (u)

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Ionizing events can be measured by many basic methods and most of them are used in various types of radiation instrumentation. Probably the most important method is measuring the ion pairs formed in a gas by the passage of radiation through that gas. Such instrumentation measure either the dose delivered over the time of interest or the instantaneous dose rate. (u)

10.4.2.1 Gas-filled detectors

One type of gas-filled detector is the ionization chamber. A volume of gas is enclosed between two electrodes as illustrated in Fig. 10.4:2. A potential of a few hundred volts is established across these electrodes, and an electric field is set up in the volume occupied by the gas. Ionization in this gas produces ion pairs, i.e., positively charged atoms and negatively charged electrons; and under the influence of the electric field, the charged halves of the ion pair migrate toward the electrode of opposite sign. This results in a small instantaneous pulse of current for each ionizing event. During the migration of ions, there will be some recombination and resulting loss of collection current. However, by increasing the strength of the electric field sufficiently it is possible to collect essentially all of the ion pairs produced within a given volume. If this volume were exactly one cc or 0.01293 g of air, one could measure the number of ions produced and obtain directly an absolute measurement of the dose in roentgens. (S)

The air cavity can be surrounded by a volume of air or air-equivalent plastic so that a necessary equilibrium is established between the secondary electrons formed within the measured volume which leave it before being measured, and those which are formed outside the volume and enter it and are measured. Also by surrounding such a volume with a tissue-equivalent plastic, proper interpretation of the observed current can be used to obtain a measure of the actual dose that is delivered to tissue. A detector based upon this principle is referred to as an ion chamber since it measures the actual number of ionizations and electronically translates this into units of dose or dose rate as shown by Hine, et al. (S)

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Another type of gas-filled counter is the proportional counter. One of the electrodes in Fig. 10.4:2 consists of a very small diameter wire and the other electrode consists of a cylindrical conductor which surrounds the wire. The electrical field established is relatively weak farther from the wire and relatively strong close to the wire, and theoretically goes to infinite field strengths for an infinitely small wire. Under these conditions an ionizing event in the gas will result in the halves of the ion pairs being separated at an increasingly rapid rate due to the stronger field near the center wire, and as the electron approaches the wire it becomes highly accelerated. When the electron reaches sufficient velocity it collides with atoms of gas, and creates additional ion pairs. This results finally in an avalanche of a large number of ions as compared with the number originally produced. This phenomenon, known as gas amplification, is used in a number of radiation detection instruments. (S)

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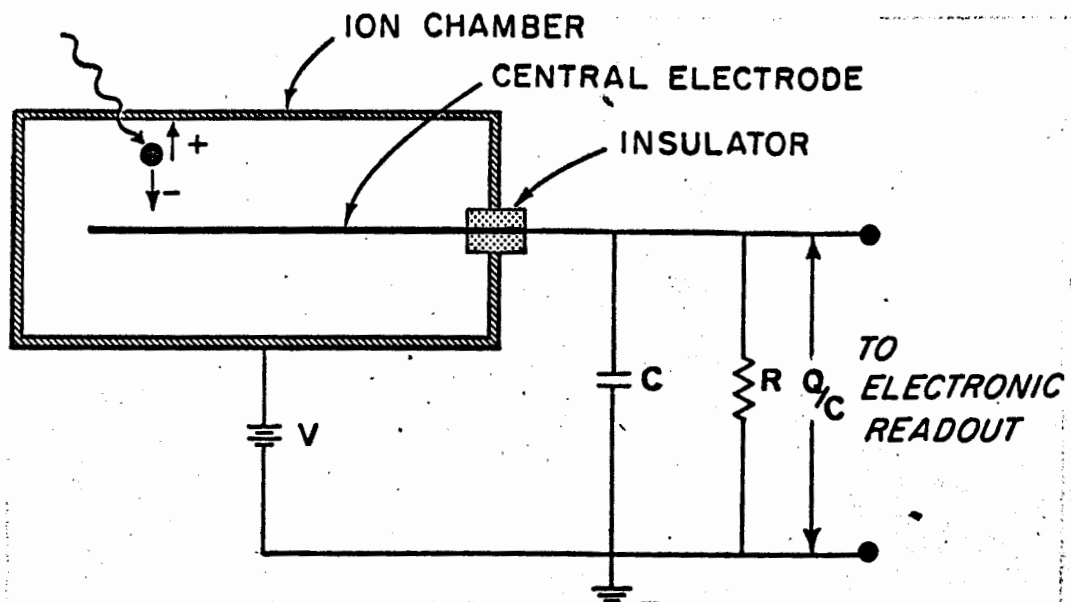
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Fig. 10.4:2 Schematic Diagram for Pulse Operation
of a Gas Filled Detector

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$R = 1.10 \times 10^4$

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If the chamber is operated with the proper type of gas and electric field strength, the degree of gas amplification is constant, and for every ion pair formed in the gas a fixed number, perhaps 10 to 100 thousand ion pairs will be collected at the electrode. When highly ionizing radiation, such as an alpha particle, passes through the gas it creates a relatively large number of ion pairs in a given path length and a correspondingly greater signal results than is produced in an ionizing event such as that caused by interaction of a gamma photon. If the chamber is connected to an electronic system which will bias against small pulses of current, one can discriminate against gamma radiation and obtain a measure of the alpha radiation alone. By proper adjustment of the voltage, a similar discrimination can also be obtained between beta and alpha particles. (S)

One of the earliest types of gas-filled counters to be used is the Geiger counter which operates on the basis of a somewhat different phenomenon. If the electrical field is increased above that used in proportional counters either by decreasing the diameter of the central wire or by increasing the voltage, any ionizing event occurring at any place within the gas volume results in a shower of ion pairs. ~~The electrical discharge explodes into a sheath of ions along the central wire and the current collected by the wire is of the same magnitude whether the sheath was initiated by a single ionizing event or by a large number of ion pairs.~~ (S)

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In summary, electrodes properly placed in a volume of gas can be used to measure directly the effect of radiation, i.e., ionization in the gas. Radiation dose can be measured by collecting only the ions produced by the ionizing event. Different types of radiation can be selectively measured by utilizing limited gas amplification (proportional region), or alternatively every ionizing particle interacting in the gas can be counted by operating in the Geiger region. Use of these basic phenomena is made in the design and operation of an extremely wide variety of radiation detection equipment. However, the fundamental principle of all ionization chamber instruments is based upon one of the three mechanisms outlined above. In all these instruments the effect of ionization in a gas is eventually converted into an electrical signal capable of being read on a suitably calibrated meter or other readout device. (S)

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10.4.2.2 Scintillation detectors

Certain materials such as inorganic crystals, organic solids, and solutions transform the energy released by ionizing radiation into a tiny burst of light in the visible or ultraviolet spectrum. Such materials are known as scintillators or scintillation phosphors.

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Scintillation counting was one of the first mechanisms used for radiation measurement. With the development of the photomultiplier tube, the use of scintillation detection has been greatly extended. (X)

With proper detector design, the flashes of light from the scintillator are proportional to the total energy deposited by ionization in the scintillator by the incident particle or photon. These light flashes, although very small, can be observed by a light-adapted eye. However, if such a scintillating material is connected to a photomultiplier, the light pulses may be converted into an electrical signal which then can be read out on a suitable device with much greater sensitivity than the human eye. The scintillation system has an advantage over a gas-filled system in that there are many more atoms in the denser crystals with which the radiation can interact. In the case of gamma radiation which interacts only rarely, this increased density greatly increases the probability of interaction and, thereby, increase the sensitivity of detection. (X)

10.4.2.3 Film dosimeters

Ionizing radiation can also be measured by its interaction with a photographic emulsion containing silver, similar to ordinary photographic film. Subsequent development and fixation of the exposed emulsion results in blackening of the developed film approximately in proportion to the degree of ionization which occurred within the emulsion. Photographic emulsions may also be used for observation of individual particles of highly ionizing radiation such as alphas, fission fragments, and cosmic radiation. It is possible to observe with a microscope the "tracks" left by the passage of a single ionizing particle through a sheet of film (Fig. 10.4:3). In this way energy absorption in a solid can be observed (X).

10.4.2.4 Solid-state and chemical dosimeters

In recent years various solid-state devices for measurement of ionizing radiation have been developed. The most promising of these are the semi-conductor devices which utilize the ionization produced in a crystal to establish an electrical signal. These devices are analogous to a solid-state proportional counter in that the size of the pulse produced is proportional to the energy deposited. Other solid-state effects such as modification of fluorescent or thermoluminescent properties, degradation of plastics, or increased conductivity of solid materials in a radiation field have also been exploited to provide a means of radiation dosimetry. (X)

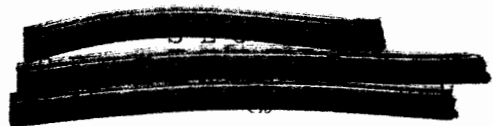
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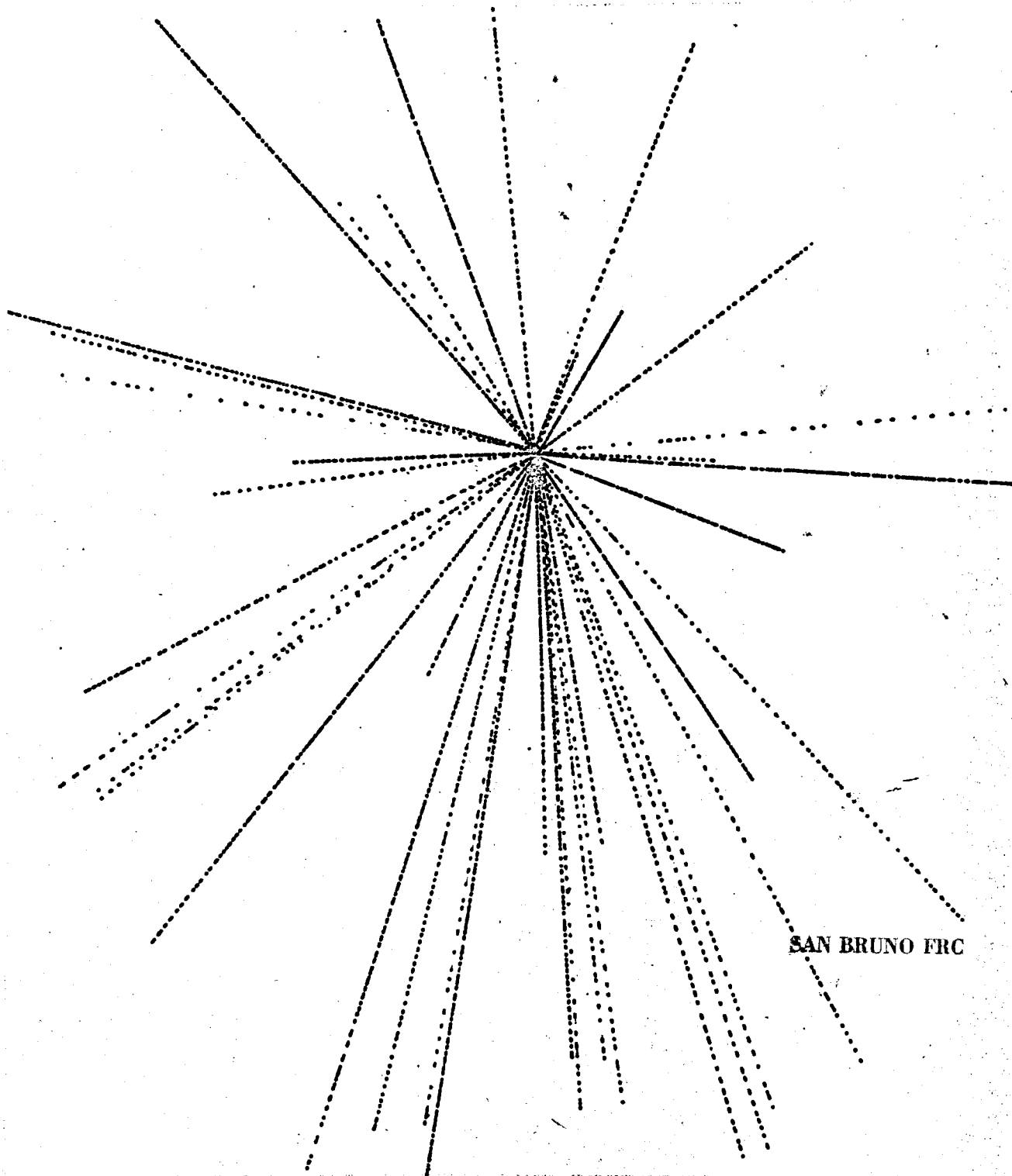
Another system coming into more general use is that of chemical dosimetry where chemical reactions resulting from ionization are used to provide an indication of radiation dose. An example is the ferrous-ferric dosimeter discussed by Price, in which an oxidation of ferrous ion in solution to the ferric state is used to measure doses of radiation. (X)

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Fig. 10.4:3 Application of Film Dosimetry and Nuclear Emulsion Technique
Showing "Tracks" of Individual Particles

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10.4.2.5 Cloud and bubble chambers*

These two methods evaluate radiation by means of the visible path produced by an ionizing particle in a chamber containing unstable gas or liquid. Activation of condensation centers in the chamber medium results in the formation of visible bubbles or droplets along the particle's path. Tracks of particles in both the bubble and cloud chamber are generally photographed for evaluation. Both of these devices are generally limited to use with highly ionizing particles such as those found in particle accelerator work, and this method of detection ordinarily is not utilized in routine radiation instrumentation. (N)

10.4.2.6 Calorimetric dosimeters

Since radiation dosimetry is essentially a measurement of energy deposition, it is possible to measure dose or dose rate by micro-calorimetric methods through the increase in temperature of a material exposed to radiation. However, such methods are normally limited to specialized laboratory use since the temperature increase is small ($.001^{\circ}\text{C}$ or less) for ordinary doses. Calorimetric methods are used, however, to obtain absolute calibration of radiation fields. (N)

10.4.2.7 Neutron dosimeters

As indicated earlier, neutrons, due to their lack of charge, are considered as ionizing radiation only by virtue of secondary rather than primary interactions. These include scattering and capture reactions, and both of these phenomena are used in the measurement of neutron fields. Proton recoils from elastic scattering of neutrons may be produced in ionization or proportional chambers composed of plastics or other material containing large amounts of hydrogen. The ionization which results from the recoil protons is measured. Neutron capture is often utilized in a counting system loaded with the enriched isotope of boron, B^{10} , which has a high capture-cross-section for thermal neutrons. When a neutron is captured in boron it immediately releases a highly ionizing alpha particle. When the boron is present as a gas, such as BF_3 , or as a solid that is coated on the inside surface of a proportional counter, neutrons can be measured by the indirect measurement of the resulting alpha particles. (N) SAN BRUNO FRC

Neutron capture is also utilized in numerous foil-activation techniques where a piece of material such as Au or S is placed in a neutron field for a given period of time and allowed to become radioactive. The induced radioactivity of the foil is measured and the incident neutron radiation calculated. By proper choice of the elements to be activated, the approximate energy spectrum of the incident neutrons can be determined. (N)

10.4.2.8 Biological dosimeters

Ionizing radiation results in biological damage which is large in proportion to the amount of energy deposited. One effect of interest is produced by chemically active entities called "radicals" which are formed by ionizing radiation and which react readily to produce various chemical changes such as the denaturation of proteins, splitting of large molecules, inactivation of enzymes, and the formation of toxic materials such as peroxides. Radiation results in damage or destruction of individual cells and, in sufficient quantity, results in damage to the organism as a whole or in death. Damage is also incurred through effects on the hereditary material contained in the cell nucleus which may not be evident for many generations of the cell following the initial irradiation. These effects are basically biochemical in nature and may be used as an indication of radiation dose through measurement of effects on cell cultures, micro-organisms and animals including man. (A)

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